Universitat Autònoma de Barcelona

Earliest pottery uses in north-eastern Iberia.
Organic residue analyses on vessels from the first farmers and herders

Adrià Breu Barcons

Earliest pottery uses in north-eastern Iberia

Organic residue analyses on vessels from the first farmers and herders

PhD Thesis

Author: Adrià Breu Barcons
Supervisors: Anna Gómez Bach
Miquel Molist Montaña
Antoni Rosell Melé
Tutor: Miquel Molist Montaña

2019
Doctorat en Arqueología Prehistòrica
Departament de Prehistòria
Facultat de Filosofía i Lletres
Universitat Autònoma de Barcelona

To the future reader, may
this tiny piece of the past
enlighten your present.

Aknowledgements

It would be deeply misleading to say that this PhD thesis is solely the result of the author's work. On the contrary, it would have not been possible unless archaeologists and chemists would have come together to bring light to our common past. Theirs is the merit of this research too.

Joining analytical science and history is not an easy task, this is why I must sincerely thank my three co-superivsors, Anna Gómez, Miquel Molist and Toni Rosell for their help and guidance throughout the process. Their teachings not only on how to bring forward a thesis but also on the development of sound research has been most valuable. Members of the academic staff of several institutions have also provided insights and contributed with their opinions. Maria Saña, Joan Antòn Barceló, Roberto Risch, Rafael Micó, Xavier Clop, Carl Heron, Ben Stern, Rebecca Stacey, Diego Tamburini, Joanne Dyer, Martine Regert, Alexandre Lucquin, André Colonese and Oliver Craig are deeply thanked for their thoughts and conversations about the insights of Organic Residue Analysis.

This PhD project has also been possible thanks to the support of many archaeologists and their tireless work in the field. Isabel Pereira, Oriol Granados, Robert Farré, Javier González, Karin Harzbecher, Ferran Borrell, Carles Tornero, Oriol Vicente, Ferran Antolín, Manel Edo, Pablo Martínez, Xavier Esteve, Xavier Oms, Josep Mestres, Juan Ignacio Morales, Artur Cebrià, Marc Piera, Josep Maria Vergès, Marta Fontanals, Josep Bosch and Maria Àngels Villalbí must be deeply thanked for the opportunity to work with archaeological materials from the sites they exavated.

Analytical chemistry is the result of team work. Beatriz Bastos, Belinda Hill, Anu Thomson, Christopher Musell, Blandine Courel, Kevin Gibbs, Ferran Colomer, Pau Comes, Elena Molina, Nadia Tarifa, Nuria Moraleda, Emili Revilla, Montserrat Pugés and Ramón Álvarez selflesly helped in the analysis of many of the samples presented in this research. Sharing the experience and challenges of bringing forward a PhD research was also shared with other fellow PhD students: Joaquim Sisa, Andreu Monforte, Ivan Gironès, Silvia Calvo, Ricard Arnaiz, Roger Alcàntara and Maria Raja.

Stages in the University of Bradford and The British Museum over the course of these years helped forge bonds with many friends who, in their own way, also supported and helped push this project forward. Patxi Ramallo, Sanne Boekenkast, Akshyeta Suryanarayan, Sam Harris, Edward Standall Aripekka Juno and many more made the journeys abroad feel like home.

Finally, this PhD would have not been possible without my chidlhood friends my family and my partner. Alex Congost, Aaron Borrego, Irene Mateo, Eloi Breu, Jordi Breu and Adela Barcons stood by my side in the best and in the worst moments.

To all of them and many more, thank you very much for your time, for your opportunities and for your patience but, overall, thank you for making a difference.

Abstract

The appearance and spread of the Neolithic way of life constituted an event which profoundly affected humankind. Research on the economic changes of the shift from hunting and gathering to agriculture and pastoralism suggests that culinary practices would have been equally affected. Beyond the fulfilment of a biological need, eating is a social act whose study offers the opportunity to gain insights into the relationship between the economic and cultural spheres of society. A new wave of research is revealing new data on the possible culinary practices in the early Neolithic across Europe, but the western end of the Mediterranean neolithisation path remains poorly understood. The idiosyncrasies of its abrupt maritime landscape provided a number of opportunities for the newly arrived farmers and shepherds to tune their economic strategies to this new territory. Therefore, this PhD research aims at evaluating the relevance of terrestrial and marine animal and plant products within the Early Neolithic culinary practices by selectively exploring culinary techniques facilitated by the appearance of pottery as a cooking tool and foodstuffs which may be linked with the landscape: marine resources and ovicaprine secondary products. To this end, we have chemically characterized and evaluated with bayesian mixing models the occurrence of lipid distributions embedded in pottery from 14 archaeological coastal sites across the northeast of the Iberian Peninsula.

A total of 114 Cardial and 76 Epicardial pottery fragments have been analysed, of which 82% yielded lipids most probably originating from degraded animal fats. Although the search for specific plant and marine biomarkers did not yield significant results, samples containing evidences of food heating beyond 250ºC suggest that the presence of cooking practices which would have reached temperatures higher than those in boiling or stewing. Furthermore, compound-specific carbon isotopic analyses have revealed that the presence of ruminant dairy and non-ruminant adipose fats in archaeological sites is mutually exclusive. Contrary to the case in the central Mediterranean, pottery in caves from the northeast of the Iberian Peninsula did not present higher quantities of ruminant dairy residues than its open-air counterparts. Alternatively, a positive correlation could be detected between the ovicaprine archaeozoological record and the intensity of the dairy isotopic signal. This evidence supports the existence of a shift from cattle milking in the Middle East to the additional secondary management of other species such as sheep or goats in the Mediterranean. Moreover, the lack of association between pig remains and non-ruminant adipose signals implies the possible existence of culinary practices in which pottery was not involved. In conclusion, data from this region supports the notion of Mediterranean-specific culinary practices from at least as early as the first Neolithic.

As a consequence of this research, it becomes increasingly more relevant to develop additional analyses capable of differentiating dairy residues at the species level. Additionally, further studies on non-edible organic residues including conifer resins may be able to inform the additional uses the first pottery in the Iberian Peninsula might have had.

Resum

L'aparició i difusió del mode de vida neolític constituí un esdeveniment que afectà profundament les societats humanes. La recerca centrada en els canvis econòmics del pas de la caça i la recol·lecció a l'agricultura i la ramaderia suggereix que les practiques culinàries es podrien haver vist igualment afectades. Menjar és un acte social que transcendeix la satisfacció d'una necessitat biològica i, per tant, el seu estudi ofereix una oportunitat única d'obtenir noves dades sobre la relació entre les esferes econòmiques i culturals de la societat. Una nova onada de recerca està revelant noves dades sobre les possibles practiques culinàries practicades a l'inici del neolític europeu, però l'oest de la branca mediterrània de la difusió del neolític, la Península Ibèrica, encara no està prou ben estudiada. Les característiques de l'escarpada costa mediterrània oferiren a les primeres societats agrícoles de mitjans del VI a mitjans del V mil·lenni Cal BC l'oportunitat de modificar lleugerament les seves estratègies econòmiques per adaptar-se a aquest nou territori. En conseqüència, aquesta tesi doctoral ha avaluat la rellevància de productes vegetals i animals en les pràctiques culinàries del neolític antic explorant selectivament tècniques culinàries més fàcilment practicables gràcies a l'ús de la ceràmica i aliments possiblement lligats al tal paisatge: productes secundaris d'ovicaprins i recursos marins. Amb tal fi, hem caracteritzat químicament i avaluat amb models bayesians l'existència de lípids conservats dins matrius ceràmiques en 14 jaciments costaners del nord-est de la Península Ibèrica.

Un total de 114 fragments ceràmics Cardials i 74 d'Epicardials han estat analitzats i un 82% han retornat lípids molt probablement resultat de la degradació de greixos animals. Tot i que la cerca de biomarcadors específics per a aliments vegetals i marins necessitarà encara d'un treball més profund, mostres presentant evidències de menjar escalfat a temperatures superiors a 250°C suggereixen l'existència de pràctiques culinàries que impliquen temperatures més altes de les necessàries per bullir o estofar. A més, l'anàlisi isotòpic de compostos específics ha revelat que l'existència de greixos làctics de remugants i greixos adiposos d'animals no remugants és mútuament excloent en els jaciments estudiats. A diferència del Mediterrani central, les coves analitzades al nord-est de la Península Ibèrica no semblen presentar quantitats majors de residus làctics de remugants comparat amb jaciments a l'aire lliure. Alternativament, s'ha detectat una possible correlació entre la quantitat de restes d'ovicaprins i la intensitat de la senyal làctica als residus. Aquest fet dona suport a l'existència d'un canvi respecte l'estratègia d'obtenció de la ll*et al.* Proper Orient, probablement centrada en els bòvids, i reforça el paper d'altres espècies més ben adaptades a les condicions del mediterrani. Addicionalment, la no associació entre el nombre de restes de porc i els residus adiposos d'animals no remugants implica la possible existència de pràctiques culinàries on els atuells ceràmics no hi estarien involucrats. En conclusió, les dades d'aquesta regió donen suport a la possible existència de pràctiques culinàries específiques del mediterrani com a mínim des del primer Neolític.

Com a conseqüència d'aquesta recerca, queda palesa la importància de poder desenvolupar anàlisis addicionals capaços de diferenciar els residus làctics a nivell d'espècie. A més, aprofundir en l'estudi de residus no comestibles com la resina de pi permetrà conèixer altres rols que la primera ceràmica de la Península Ibèrica hauria pogut tenir.

Table of contents

List of figures

List of tables

1 Introduction

A considerable amount of recent research (PhDs and published papers) has undertaken the task of providing comprehensive synthetic views of the distinct material evidences hitherto recovered from the Early Neolithic in the north-east of the Iberian Peninsula. Studies on lithic tools (ex: Borrell 2008, Borrell and Gibaja 2012, Palomo 2012, Gibaja *et al.* 2018a), plant remains (ex: Allué 2003a, Antolín 2013, Buxó 2016, Revelles 2017, Piqué *et al.* 2018), animal remains (ex: Miró 1991, Colominas *et al.* 2008, Saña *et al.* 2015, Nadal 2016, Navarrete 2017) and pottery and pottery decorative styles (Blasco *et al.* 2005, Bosch *et al.* 2011, Oms 2014, Gómez 2016) have been complemented by the publication of monographs focused on specific regions and archaeological sites (el Pla de Barcelona: Molist and Gómez 2016, La Draga: Bosch *et al.* 2000, 2011) and frequent gatherings where research is presented amongst peers (ex: Taula rodona de Montserrat, Congreso del Neolítico peninsular, Workshop Impressa, specific sessions in international congresses such as UISPP or EAA).

Thanks to these collective efforts, the available data for the Early Neolithic period in the north-east of the Iberian Peninsula is reaching an unprecedented degree of detail. Because of it, previously unresolvable questions can now be explored, which proves that this particular field of research is in a healthy and dynamic state despite of all the limitations and shortcomings of modern day academic research.

The amount of knowledge gathered and available nowadays compared with what was at hand at the beginning of the twentieth century is strikingly high. Moreover, it could be argued that the field is now well beyond a state of basic description and has already entered the next phase in research based on the formulation and falsation of social hypotheses.

In a period where archaeologists specialised in the study of many types of remains have already made significant field-changing contributions, attempting a wide synthesis could easily fall into oversimplifications and the misinterpretation of results given the nuances and limitations of each specific speciality.

Nevertheless, it is also true that the reader and the results of the following research must first be put in a proper context which highlights its relevance. The following pages are an attempt to provide an updated overview of the socioeconomic reality in place between the years 5600 and 4500 cal BC in the north-east of the Iberian Peninsula.

Agriculture and pastoralism in this region might have been introduced by pioneering groups which have been archaeologically substantiated by a handful of impressa-like decorated pottery sherds within sites containing the ealiest Neolithic ^{14}C dates (Bernabeu *et al.* 2009, Oms 2014). Barely no further evidences of this possible pioneering stage are available nowadays and it is still a matter of significant debate whether future research will be able to effectively detect, excavate and study comprehensive archaeological contexts from this period.

Regardless of the pottery decorative styles performed by the firsts farmers and shepherds, there is a compelling and strong evidence placing domesticates within the region by the year 5500 cal BC. These would be accompanied by a package of other archaeological evidences such as macrolithic tools and pottery usually decorated with the use of toothed shells (*Cerastoderma edule* or *Acanthocardia tubercullata*) in what has been described as the Cardial technique and complemented with appliqués and incisions with other tools, which gives name to the human groups practicing it and to the overall period.

At that moment in time, the coastal region between the Ebro river and the Pyrenees mainly presented the same orogenic features with the exception of the coastline, whose deltas and beaches might have significantly varied from its present position. Although its location is well known in some areas, it has not been yet well defined in others. In essence, the Ebro and Llobregat deltas did not exist (Sornoza *et al.* 1998, Julià and Riera 2012) and the general coastline was placed differently due to the effects of the Flandrian transgression. Rather than the presently widespread conifer forests grown after severe deforestation in the medieval and modern times, pollen records from a range of points across the territory have identified the significant presence of oaks, pines, junipers and shrubs including the lentisk, the strawberry tree and the prunus amongst many others. Within this ecosystem, the first farmers and shepherds most possibly found deer, foxes, badgers, boars, bears and wild goats grazing in plains, rivers and mountains (ex: Saña *et al.* 2015, Miró 1991).

The first settlements in the Cardial period most probably involved the construction of huts and enclosures for the herds with primary and secondary buildings (Guixeres de Vilobí, La Draga, Barranc d'en Fabra, Barcelona's Raval) and the opening of forest clearings to grow domesticates. Rather than a slash and burn unintensive strategy, paleobotanic remains suggest that these first farmers intensively exploited their fields possibly by manuring and applying the necessary care to obtain sufficient yields to fill an increasing amount of underground silos which would be later used to dispose of waste. This type of agriculture would make sense in a world with large scale forests and would trigger the firsts significant deforestations (Riera 1994). The abundant cavities in such a calcareous landscape would have been quickly exploited as shelters for herds and sacred places where to bury the dead (Can Sadurní, Cova de l'Avellaner, Cova Bonica and Cova Foradada)(Gibaja *et al.* 2018b, Oms *et al.* 2017), although exceptional open air burials (Plaça Vila de Madrid, Barcelona) have also been detected. Sheeps and goats would have been amongst the main species in the first herds, but, in several sites (La Draga, Carrer Reina Amàlia) human groups invested in cattle to obtain their meat and dairy proteins as well as animal traction. Raising pigs might have involved both closed estabulation and/or free grazing and patterns of transhumance might have mobilised sheeps and goats between the plain and the mountain. Rather than being fully abandoned, hunting and gathering activities were complementarily practiced to obtain other sources of food. Hazelnuts, acorns, pinenuts, as well as poppy seeds were recovered from Early Neolithic sites (La Draga, Guixeres de Vilobí). Bones from wild animals such as deer, wild goats, foxes or badgers were present in many human enclaves, but one of the most popular ones might have been rabbits. Lithic points and a complete bow from La Draga are exceptional evidences from the occurrence of hunting practices.

The degree of stylistic and ritualistic evidences detected in Cardial groups across the region suggests that those were rather a homogenous society with a significant degree of social integration capable of exchanging ideas and products. The characteristic red jasper from the Montjuïc mountain, near modern day Barcelona, could be found across the territory, but potters usually preferred local clays to model their vases and would have seldomly moved them away from their production area. Bracelets, pendants, rings and a spread of jewellery items were carefully shaped out of seashells and other materials such as animal bone, antler or calcite amongst others (Oliva 2015).

The diet would have most probably involved a combination of the yield from domesticates and the capture of marine and terrestrial wild resources although its specific weight is still unclear. Culinary practices transforming the original natural products have been identified in the form of dairy fats amongst pottery residues in the sepulchral layers in Can Sadurní and posterior occupations related to a non-mortuary use of the cavity.

There are no evidences supporting the existence of a division of labour due to gender or status differences, but no compelling case has been made otherwise either. Although surplus and food storage has been placed as the origin of social inequalities, no persuasive studies have been performed to assess the social power structures which might have been in place at the onset of the Neolithic in the north-east of the Iberian Peninsula.

The start of the 5th millennium cal BC marked the decline of the Cardial groups. The use of shell impressions in pottery dissipated and sticks and fingers were used instead to recreate simmilar decorative patterns. By the second quarter of the 5th millennium the impressions had practically disappeared and the use of appliqués and/or incised decorations had become widespread. This change in decorative practices was used by archaeologists to mark the end of the Cardial period and the beginning of the Epicardial (Martín *et al.* 2010) horizon. In fact, not only decoration changed, but shapes modulated and clay tempers shifted from grog to calcite (Clop 2011).

Although the so called Epicardial moment might be part of a second Early Neolithic in the eastern coastal areas of the Iberian Peninsula, it is truly the marker for the arrival of agriculture and pastoralism more inland. As seen in Figure 1, there is a clear lack of Cardial sites in the internal regions of modern day Catalonia whereas Epicardial sites have been located in a wider territory (Mestres 1992). It seems that, although the first Neolithic societies did not immediately expand across the territory, the general trend is that its new economic system would become the new norm. Therefore, these set of partial transformations indicate that some degree of social change was in motion. It has been suggested that, although it was considered vital that the shell was involved in the Cardial decoration process, this indispensability disappeared in Epicardial pottery (Martín *et al.* 2010). This would correlate with a world where Neolithic communities would have been spreading inland (Mestres 1992).

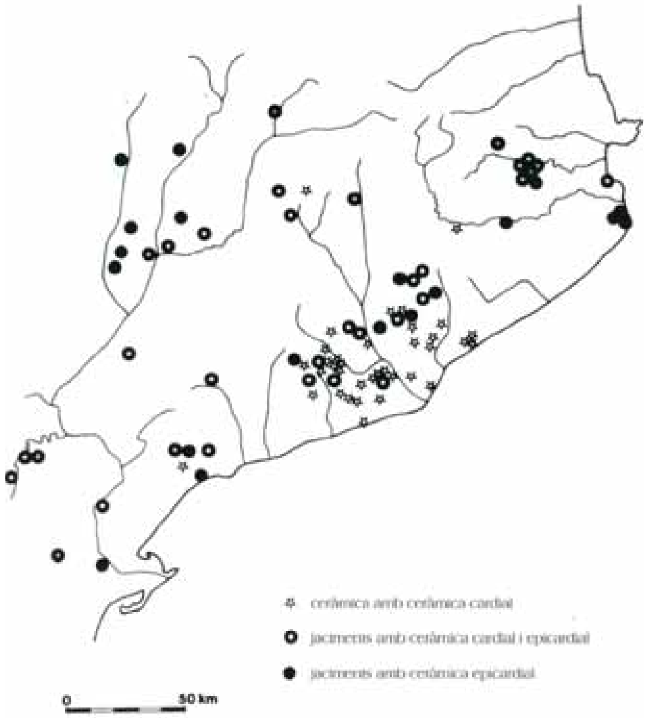


Figure 1: Map showing the distribution of Cardial and Epicardial sites across the northeast of the Iberian Peninsula (modern day Catalonia). Reproduced from Mestres 1992.

It is also important to note that economic data from both Cardial and Epicardial archaeological contexts are very similar. Carpological analysis from sites across the modern Catalan territory confirm regional differences within, but show no variation between the second half of the VIth (Cardial) and the first half of the Vth (Epicardial) millennia cal. BC (Antolín 2013). The creation of new agricultural fields would have come at a cost and, in fact, by the end of the 5th millennia cal. BC forest degradation caused by anthropic pressure started to clearly appear in the pollen records (Martín *et al.* 2010). In terms of the zooarchaeological evidence, the management of herds varied more geographically than chronologically in the early north-east Iberian Neolithic.

Rather than changing the economic system, societies in the Epicardial period continued practicing agriculture and pastoralism, although pottery decoration and temper types (Blasco *et al.* 2005, Oms 2008, Clop 2011) suggest that some degree of social change did happen. In essence, the first Neolithic societies in the Iberian Peninsula had the chance to modify its economic system. They could have certainly been in contact with hunter-gatherer societies inland (Bernabeu 1999), but they preferred to maintain their own status quo to a degree. If pottery is one of the few indicators of social change, therefore, research towards comprehending its uses is key to understand the mechanisms that promoted the strengthening of the agro-pastoral economical system.

In the spirit of this line of academic research, the next section will provide a historic overview of the main social hypotheses in place behind the study of Early Neolithic pottery since its first detection at the beginning of the XXth century, to the present day. It is the intent of this exercise to reflect on the importance of detecting research opportunities which significantly increase the value of pottery as an indicator of the social practices in place at the onset of the Neolithic. It is important to notice that the research since the identification of the fist cardial wares until the most recent syntheses has seen different theoretical approaches applied to this pottery type. Therefore, what follows is a more in depth look at cardial pottery as a chronocultural marker, a Neolithic marker, as an artistic phenomenon and, possibly, as a tool.

1.1 History of the research on the Early Neolithic in the northeast of the Iberian Peninsula.

Starting at the origins, the first identification of cardial pottery was performed on material from a set of caves in Montserrat (Cova Freda and Cova Gran) by Colominas (1925). In this first publication, the material found in the sites was briefly described as pottery which had been decorated using a Cardium edule seashell. The research of similar artefacts barely gave any singificant results, as Colominas could only locate six fragments in three additional caves: Cova de Can Pascual (Castellví de la Marca), L'Espluga Negra (Castelltort) and Balma de la Llera (Lladurs). Nevertheless, the singularities of these artefacts were used by the author to name a new archaeological horizon, the Monsterratine culture (Figure 2).

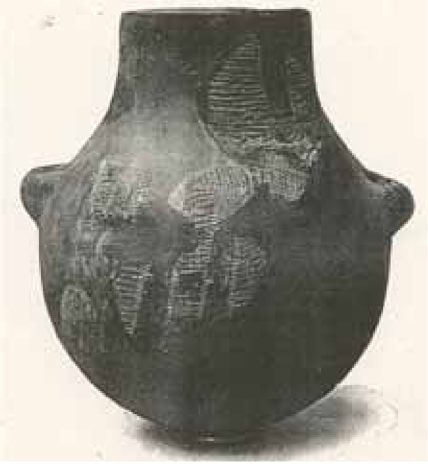


Figure 2: Vessel deocrated with cardial incision from Cova Freda (Colomines 1925)

Furthermore, when trying to assess its possible age, Colominas noted the similarities between cardial wares and bell beakers and suggested that the two phenomena were close in time. The construction of a full “cardial period” was, therefore, not possible until further research was carried by M. Grivé (1931) when excavating l’Esquerda de les Roques del Pany. The stratigraphy preserved in the cave evidenced that cardial wares were in significantly lower layers than the bell beaker potsherds. Nevertheless, the fact that a metal object was also found within the cardial layers led the archaeologist to conclude that the “Monsterratine” culture was placed in a transition moment between the Neolithic and the Chalcolithic (then named Eneolithic).

After this initial research before the civil war (1936-1939) and before the 1950’s, new sites containing cardial wares were reported by local experts. Dr Vilaseca (Vilaseca 1945, Vilaseca and Mercadé 1957) possessed an wide collection of Neolithic pottery (amongst which cardial fragments could be found) which was used by the italian archaeologist Luigi Bernabó Brea, excavator of Arene Candide, to identify the cardial wares as the distinct pottery type marking the neolithisation horizon in the western Mediterranean (Bernabó 1956). The work of Dr. Vilaseca culminated in 1973 with the publication of “Reus y su entorno en la Prehistoria”, where several sites containing cardial pottery were described. Furthermore, in this same decade, a new wave of archaeologists excavated and reported new sites which fitted this horizon. These were Roc d’en Sardinyà (Baldellou 1972), Cova del Pèlag (Virella 1973), Cova Bonica (Baldellou 1974), Cova de la Valldany (Cura *et al.* 1979) and Balma de Llera (Rovira 1976). With the arrival of the first ^{14}C dates, the hypothesis that the cardial horizon belonged to the first Neolithic societies in the western Mediterranean was confirmed. (primera datació) Cova del Parco was the first site from which radiocarbon dates were published (Alonso *et al.* 1978) and Cova del Toll (Guiliane 1979) quickly followed with a range of their own. In consequence, when in 1981 the archaeologists working in the Early Neolithic at the northeast of the Iberian Peninsula met at the Monsterrat round table, eleven ^{14}C dates belonging to the Early Neolithic and originating from Cova del Parco, Cova del Toll and Cova de la Font del Molinot were already available. Therefore, absolute dating from that time fully supported the proposal that, at the moment of implantation of agriculture and pastoralism, pottery was decorated per the stylistic cardial rules.

In summary, in the first 50 years of research (aprox. 1925-1975) cardial wares journeyed from a scarcely found and chronologically unclear decoration type to a marker for the origin of the Neolithic across the territory. These results still stand nowadays, even if the cardial technique is essentially a seashell used as a tool to mark wet clay. For the next 20 years new sites containing cardial wares were excavated and, due to new ^{14}C dates or stylistic comparison, the corpus of this pottery type significantly grew. At this time, the discussion shifted to the characterisation of the arrival of the Neolithic and how it would have permeated the territory.

The 1980's marked a period of profound development of the archaeological practice which reached archaeologists working on the Early Neolithic in the northeast of the Iberian peninsula. Examples of this are Cova del Frare (Martín *et al.* 1981, Martín *et al.* 1984), Guixeres de Vilobí (Mestres 1981), Can Banús (García *et al.* 1982), Can Soldevila (Costa *et al.* 1982) and Pedrera de Sant Jordi (Gràcia 1987). Moreover, the influence French authors such as J. Guilaine would have on the development of the archaeological research must be mentioned. Works such as "Prémiers bergers et paysans de l'Occident Méditerranéen" (Guilaine 1976b), "La neolitización de las costas mediterráneas de Francia y España (Guilaine 1976a) and "Le Néolithique ancien en Languedoc et Catalogne, elements et réflexions pour un assai de périodisation" (Guilaine 1984) were amongst the most influential.

Important milestones in this moment were the publication of the round table celebrated in Montserrat (1981) and the formation of the devolved catalan institutions after the transition from Franco's dictatorship to democracy. The publication of "Les excavacions arqueològiques a Catalunya en les darrers anys" by the Department of Culture of the Generalitat de Catalunya published and brought to light a significant quantity of diverse sites, some of which also contained cardial wares (Cova dels Dos, Cova de les Ànimes and Balma de l'Espluga). Moreover, this new impulse from the 1980's culminated in 1991 with the celebration of the 9th International Col·loquium of Archaeology in Puigcerdà, which, that year, focussed on the state of the research on the Neolithic in Catalonia. In this meeting, cardial ware was definitively confirmed as the marker for the Early Neolithic and new synthetic and interpretative proposals looking at the arrival of the Neolithic phenomenon through pottery were laid out (Martín 1992, Mestres 1992).

Moreover, new archaeological sites dating from the period were also presented in this congress. They were Pla de la Bruguera (Fíguls 1995) and Plansallosa (Alcalde *et al.* 1991). In 1995 reports from the excavations in Barranc d'en Fabra (Bosch *et al.* 1995a, 1995b) and a monograph volume on Balma Margineda (Guilane and Marztluff 1995) was published. Moreover, in 1996 an in depth study of materials from Cova del Parco (Petit 1996) was presented. This coincided with the celebration of the first Congres of the Neolithic in the Iberian Peninsula. Their proceedings were a new platform where to report data from recently excavated sites such as Font del Ros (Bordas 1995) and, in its second edition, Can Sadurní (Blasco *et al.* 1999), La Draga (Bosch *et al.* 2000) or Balma del Serrat del Pont (Alcalde 1999). If a common denominator should be found linking all these studies from the end of the XXst century and the beginning of the XXIst it would be easily found that pottery analyses were surpassed as the core line of research into the early cardial Neolithic. In the monographies from La Draga (Bosch *et al.* 2000, Bosch *et al.* 2011), pottery studies would be next to a wide range bioarchaeological analyses. This was the perfect example of the arrival and development of a new multidisciplinary way of researching in archaeology. In consequence, it could be said that the 9th international colloquium on Archaeology in Puigcerdà could mark the arrival the British "New Archaeology" to Early Neolithic research in the northeast of the Iberian Peninsula.

Work from authors such as Fugazzola-Delpino *et al.* (2002) presenting new synthesis across the Mediterranean would now be accompanied by new monographic publications composed by the range of studies performed by different experts on Balma del Serrat del Pont (Alcalde *et al.* 2002) and Caserna de Sant Pau del Camp (Molist *et al.* 2008). It would be in this moment when excavations from contract archaeology would present its first contributions such as the excavation of Can Xammar (Pou and Martí 2005), Can Roqueta (Oliva *et al.* 2008), Ca l'Estrada (Fortó *et al.* 2005), Can Filuà (Terrats 2008) or Carrer Reina Amàlia (González and Harzbecher 2008, González *et al.* 2011). When adding the publication of even more new sites such as Cova del Vidre (Bosch 2005), El Cavet (Fontanals *et al.* 2008a, Oms and Morales 2008), new data from ongoing sites (Can Sadurní: Blasco *et al.* 2011) and new excavation projects such as Cau de les Guilles (Soler i Serangeli 2007, Soler i Serangeli 2008, Soler i Serangeli 2010) the amount of archaeologically registered material culture available from the Early Neolithic was, at the end of the first decade of the XXIst century, quite substantial. Within this framework, new works of synthesis were presented in the "LI Mémoire de la Société Préhistorique Française", which focussed on the first peasant societies in the Western Mediterranean and the structure of their ceramic productions (2010). In this work it seemed that the cardial decorative technique as a marker for the earliest Neolithic was gradually complemented by a shift towards more post-processual paradigms focused on the study of the decorative patterns and the social interpretations which may arise from them.

The second decade of the XXIst century was clearly marked by the aftermath of the financial crisis, which motivated the later governmental austerity measures affecting the performance of both academic and contract archaeology. Despite the difficulties, researchers continued to report the discovery of new archaeological sites dating from the Early Neolithic. Results from new and ongoing excavations supported by funded research projects helped deepen the available knowledge from sites such as Coves del Toll (Cebrià *et al.* 2014), La Draga (Palomo *et al.* 2011, Antolín *et al.* 2014, Palomo *et al.* 2014, Piqué *et al.* 2015), Cova Gran de Santa Linya (Mora *et al.* 2011), Cova Colomera (Oms 2008, Oms *et al.* 2008 Oms *et al.* 2014), Cova de la Guineu (Oms *et al.* 2016a), Cova de l'Avellaner (Gibaja *et al.* 2018b), Sanavastre (Mercadal *et al.* 2009), Cova del Sardo (Gassiot *et al.* 2012, Gassiot *et al.* 2015), Cova sant Llorenç (Borell *et al.* 2011 and Borrell *et al.* 2016) and Cova dels Fems (Palomo *et al.* 2018). Publications arising from excavations performed by private companies were also capable of performing significant contributions to the amount of archaeological records recovered from the period. This included the sites of Cova de la Font Major (Cebrià *et al.* 2014), Pou Nou (Oms *et al.* 2016c), La Serreta (Esteve *et al.* 2010, Oms *et al.* 2014), Berenguer de Palou (Pereira 2019), Codella (Alcalde *et al.* 2008), El Pla del Serrador (Muñoz and Martínez 2014), Vinya d'en Pau (Oms *et al.* 2014), Mas d'en Boixos (Vidal 2007, Oms *et al.* 2014), Vila de Madrid (Pou *et al.* 2010), El Molló (Piera 2010a) and Pla del Gardelo (Piera 2010b)(see Figure 3).

Nevertheless, maybe one of the most encouraging new developments from this period is the initiation of research projects in sites which had played an integral role in the definition of the Early Neolithic but had not been excavated for a long period of time. Such cases included Cova Bonica (Oms *et al.* 2017, Daura *et al.* 2019), Guixeres de Vilobí (Oms 2017a, Martins *et al.* 2015, Gibaja *et al.* 2018a, Oms *et al.* in press) and the start of a new research project in the Montserrat caves which, almost 100 years later, are being excavated again[1].

---

[1] UB press release https://www.ub.edu/web/ub/ca/menu_eines/noticies/2018/07/005.html

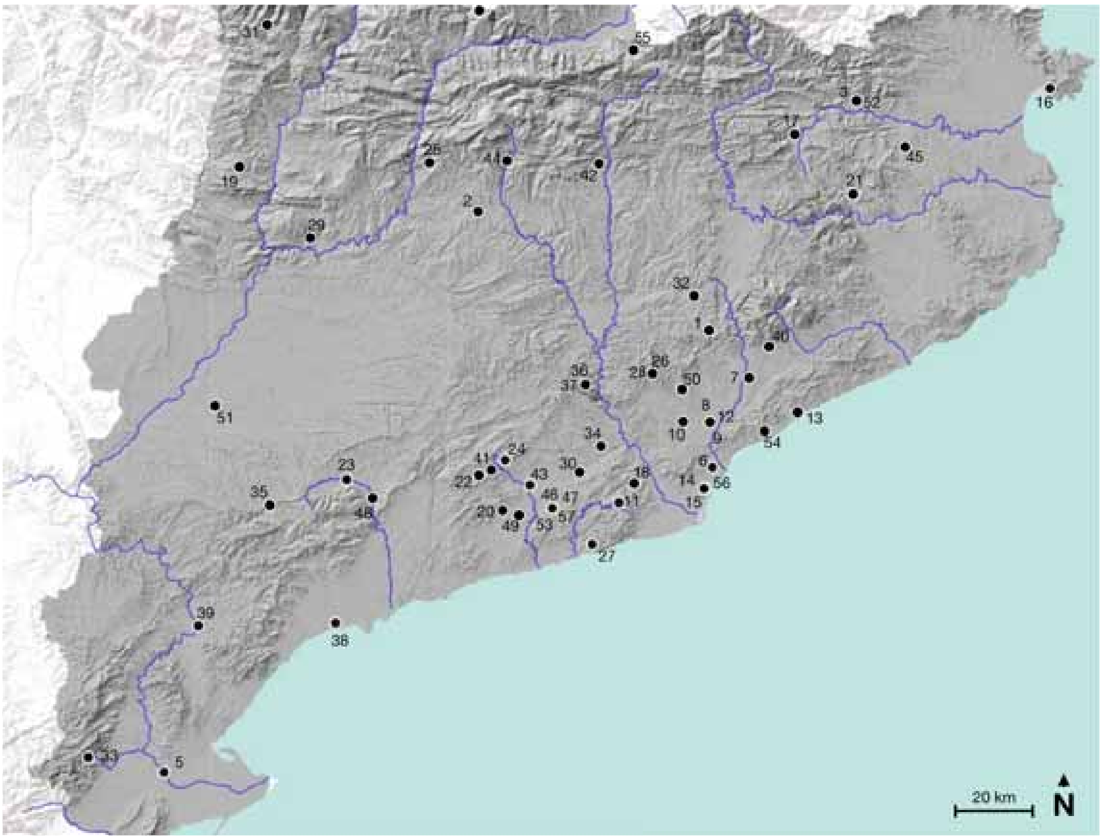


Figure 3: Map showing the approximate location of sites mentioned in the text: 1: Balma de l'Espluga, 2: Balma de la Llera, 3: Balma del Serrat del Pont, 4: Balma Margineda, 5: Barranc d'en Fabra, 6: Berenguer de Palou, 7: Ca l'Estrada, 8: Can Banús, 9: Can Filuà, 10: Can Roqueta, 11: Can Sadurní, 12: Can Soldevila, 13: Can Xammar, 14: Carrer Reina Amàlia, 15: Caserna de Sant Pau, 16: Cau de les Guilles, 17: Codella, 18: Cova Bonica, 19: Cova Colomera, 20: Cova de Can Pascual, 21: Cova de l'Avellaner, 22: Cova de la Font del Molinot, 23: Cova de la Font Major, 24: Cova de la Guineu, 25: Cova de la Valldany, 26: Cova de les Ànimes, 27: Cova de Sant Llorenç, 28: Cova del Frare, 29: Cova del Parco, 30: Cova del Pèlag, 31: Cova del Sardo, 32: Cova del Toll, 33: Cova del Vidre, 34: Cova dels Dos, 35: Cova dels Fems, 36: Cova Freda, 37: Cova Gran de Santa Linya, 38: El Cavet, 39: El Molló, 40: El Pla del Serrador, 41: Esquerda de les Roques del Pany, 42: Font del Ros, 43: Guixeres de Vilobí, 44: L'Espluga Negra, 45 La Draga, 46: La Serreta, 47: Mas d'en Boixos, 48: Molins de la Vila, 49: Pedrera de Sant Jordi, 50: Plà de la Bruguera, 51: Plà del Gardelo, 52: Plansallosa, 53: Pou Nou, 54: Roc d'en Sardinyà, 55: Sanavastre, 56: Vila de Madrid, 57: Vinya d'en Pau

To date, research on the earliest Neolithic sites in the north-east of the Iberian Peninsula continues to yield new data from both well-established archaeological projects and the inclusion of additional unpublished sites. The amount of available information and the maintenance of the research activity despite the recession offer good perspectives for the future of this field of research. As it has been previously stated, the initial development of more synthetic and transversal investigations and publications (Oms 2017b, Antolín *et al.* 2018) and its integration into the development of the first agricultural and pastoralist societies from wider geographical areas will certainly provide a wider framework in which the research of the region can be valued and can impact the study of the unique transition between the Mesolithic and the Neolithic periods.

Within the presented ecosystem of research, pottery has been one of the most resilient artefacts. It has been a valued material across the roughly hundred years since Colominas first published the first Monserratine wares, nevertheless, the motives and reasons for its importance have shifted as new and better methodological approaches entered the field of archaeology. Whether it was conceived as a chronocultural marker, an integral part of the materiality of the first farmers and shepherds or a decorative medium, pottery was, essentially, a tool. The following pages will reflect on the value of pottery for the study of the first Neolithic societies and suggest a possible way forward through the study of its uses.

1.2 The archaeological values of Early Neolithic wares

1.2.1 Chronocultural markers.

Conceptualising the cardial ware as a chronocultural marker is, somehow, the origin of the identification of this decorative technique. Since the work of Colominas (1925) until the publications of Bernabò (1956) what had been called the “monserratine” pottery was located within the Eneolithic period, a name used to define Neolithic markers deprived of a significant amount of metallic objects (Grivé 1931). In the framework of traditional archaeology the object would yield two types of information: a chronology and the movement of the populations that produced said objects. This is how the concept of archaeological culture was used by researchers in that period. The following (translated) quote from M. Grivé perfectly exemplifies the type of interpretation performed based on the presence of the cardial decorative technique:

> *“ This [cardial] culture was, later, eliminated or absorbed by the one of the bell beaker, more advanced, by acting in two different places: by the south coming up from the Millares and, by the west, from Somaén. This is the main interest of our cave, as it provides, thanks to the confirmed stratigraphy, a means to fix the chronologic position of the monserratine ceramic type compared with the bell beaker. It proves that it represents erstwhile times ended by the bell beaker.”*

Although the Cardial ware later proved to be a decorative technique from the Early Neolithic, the background theoretic structure put forward by Grivé would be in place across the XXth century: pottery informed of chronology and population movements.

Regarding chronology, the first steps towards the location of cardial pottery at the origins of the Neolithic were the ones developed by Bernabó (1956) in Arene Candide. These were based on the comparison of stratigraphic profiles and it wasn't until the late 1970's, that the first absolute dates on layers containing cardial wares could be practiced. The new radiometric methods provided the possibility of working with calendaric years obtained independently from the chronocultural classical dating strategies. This provided a means of contrasting both lines of evidence. The first radiocarbon measurements were performed by Maluquer de Motes in Cova del Parco (Alonso *et al.* 1978) (6450±230BP, 6170±70BP and 5790±170BP all three of them on an agglomerate of bones). Although in that moment they were considered valid (Marcet 1981), further publications (Petit *et al.* 1996) reviewed their contexts and concluded that there was no supporting archaeological data placing the studied bones in the Cardial or Epicardial cultural horizons. In consequence and by recommendations of the archaeologists of the cave (Petit *et al.* 1996) these dates are nowadays not treated as valid anymore.

Generally, the first ^{14}C synthesis were concentrated in limiting and sub-dividing the Early Neolithic. In fact, the scarcity of absolute dates did not allow, at first, to detail the chronological ranges. Clop *et al.* (1992) worked with 4 dates on the Cardial early Neolihtic (two already published in the Montserrat round table (Balma Margineda 6670±120, Cova del Frare 6380±310 and two from Cova del Parco 6450±230 and 6170±70). As it can be observed, the margins of error were significantly high but, for the first time, they were 2-sigma calibrated, which yielded the first hypothesis of a chronologic framework between 5750 to 4900 cal BC which, despite the increasing amount of new data and improved analytical techniques available nowadays, was surprisingly accurate when the state of the art in that moment is taken into account.

Five years later, within the framework of the First Congress of the Peninsular Neolithic (Mestres and Martín 1995), already 15 dates for the Cardial Early Neolithic were already available (the previous four and eight new ones from La Draga and single dates from Font del Ros, el Pla de la Bruguera and Cova del

Vidre). In these cases, the authors considered that the volume of dates and its margins of error wwere still insufficient to fully differentiate the three tentative Early Neolithic phases (Cardial, Epicardial and Postcardial) since these could have been one of the main causes of the apparent overlapping of its ^{14}C dates. Since then and across the 90's and 2000's new dates from Caserna de Sant Pau or Can Sadurní would be added to the growing corpus of radiometric information.

The following synthesis of available ^{14}C dates would not be performed until 2008 (Barceló 2008, Clop *et al.* 2008) within the framework of the study of the cultural sequences of all the catalan prehistory. The volume of dates taken into account would be significantly higher (559 dates for a range between 1200 cal BC to 750 cal BC). Barceló argued that the overlap between Cardial and Epicardial dates was not statistically significant, thus reinforcing the notion that they were a sequence of connecting cultural developments. The Cardial horizon was then defined between the 5400 and the 5000 cal BC while the Epicardial was considered to start around the year 5000 cal BC and last until the 4400 cal BC. In consequence, with a larger corpus of dates and significantly reduced error margins, cardial decorations were located at the second half of the VIth millennia cal BC.

As precision on the ^{14}C dates incremented (specifically thanks to the arrival of AMS dates), periods beyond the traditional cultural groups were proposed. Following Manen's work (2002) taking into account dates from Catalonia and the south of France and decorative patterns, the cardial and epicardial periods were effectively subdivided in several sub-periods. The early cardial would be placed between the 5700/5600 until the 5300 cal BC and the recent cardial would be located between the 5300 and until the 4900/4800 cal BC. This would mark the beginning of the early Epicardial, which would overlap with dates between the 5300 and 4800 and, finally, the recent Epicardial, between 5000 and 4500 cal BC. As it can be seen, the proposed ranges completely collided with the limited 400 years suggested by Barceló (2008).

The issue of the internal periodisation of the Early Neolithic would be then further explored by Martín *et al.* (2010) which proposed the existence of an early Cardial, a "Full" Cardial and a Recent Cardial which would overlap with the early Epicardial. This study used 33 dates from the cardial and epicardial Early Neolithic. The early cardial was attributed a chronology between 5800 and 5500 cal BC by using the works of Manen (2002) and the most ancient date in Balma Margineda (6670±120BP) and Font del Ros (6561±56BP). The dates from Font del Ros had a significantly constrained error margin and it originated from an extensive stratigraphic unit not constrained by any particular structure and, once calibrated (5623-5466 cal BC) it could only be placed at the end of the aforementioned early Cardial period. It must be born in mind that ^{14}C dates do not indicate a chronologic range signalling the span of human presence in a site, but rather the date of the death of the dated organism. In consequence, although the existence of an early Cardial sub-phase has been kept by the researchers, their ranges have become more constrained.

Regarding the transition of the cardial phenomenon into the Epicardial, the Cardial did not seem to go beyond the beginnings of the Vth millennia and faded away in a soft transition which contrasts with its abrupt appearance. In essence, the cardial phenomenon should be always conceived for what it really is, a decorative technique which was in fashion across a specific period in time which can, in consequence, be used to detect that said period in the archaeological record. From this point onwards, sub-phases should be used when studying the variation of the characteristic traits of the first Neolithic societies as the VIth millennia ended. In a way, Mannen's proposal linking the decorative stiles with ^{14}C dates would inspire further works.

Recently, although a phase contemporary than the cardial could be characterised by "bobquique" or "punto y ralla" type decorations at the interior of the Iberian Peninsula or the older "Sillon d'impresion (in the south of France) or "impressa" in Italy, the huge variety of names given to a range of not clearly well defined decorative techniques is still a challenge which will have to be overcome in order for this line of research to be pursued further. The detection of "impressa" type decoration in a stratigraphic position which is older than the cardial in an assemblage from Valencia (Mas d'Is: Bernabeu *et al.* 2003) opens the door to a new line of research for the VIth millennia.

Nevertheless, it should be wrong to conceive these most recent developments as the result of a traditional archaeology focused on the decoration as a cultural marker. Beyond a romantic nostalgia of the chronological value of pottery, decoration stopped being the core evidence for establishing time soon after the arrival of the first ^{14}C dates. When this notion was abandoned, the fact that pottery signalled the arrival of a range of socioeconomic changes became the source of its archaeological value.

1.2.2 Markers of the spread of agriculture and pastoralism.

The second interpretative line used by archaeological researchers has been, as previously stated, the treatment of pottery as a marker for demographic movements. Since the Neolithic diffusionist proposals by Bernabò (1956), this hypothesis was further explored by Ammerman and Cavalli-Sforza *et al.* (1988) as the accumulation of evidence which validated the diffusion of the Neolithic (the gradient of ^{14}C dates from the east to the west of the Mediterranean). The chronological markers associated with the oldest Neolithic were instead used to validate that those ^{14}C dates were, in fact, selecting the first arrival of Neolithic societies.

One the most synthetic proposals using this line of thought was the one presented by Mestres in 1992 at the 9th International Colloquium of Archaeology in Puigcerdà (Mestres 1992). This author gathered data from up to 70 sites containing cardial wares in the north-east of the Iberian Peninsula and suggested the existence of two significant regions. A: a space concentrated around the Llobregat River and close to the coast. B: the remaining of modern day Catalonia. The first region would be characterised by sites placed in the mountain ranges adjacent to the coast and to the valleys within, this would be mainly caves (80%) and also open air sites (20%) with mainly cardial decorated wares and a dense settlement pattern. The opposite would be found for region B, with a disperse settlement still mainly in caves but with a higher importance of the Epicardial wares.

Mestres (1992), using this data, suggested the neolithisation of the region occupied by modern day catalonia could be divided in three phases. In the first one, pioneers from egalitarian Neolithic societies would arrive at the coast and produce the sites found in area A. The second phase would see the expansion of said agricultural and pastoralist groups occupying area B via a reconfiguration of the cultural patterns, which would yield the Epicardial phenomenon and a reflux would bring it back to the seashore. Finally a third phase would be associated with the consolidation of these Neolithic groups in what was called the Evolved Early Neolithic (the Montboló and Molinot horizons, also known as postcardial). This proposal by Mestres is, in any case, still found within the framework of a still rather traditional archaeology whose research is based on the presence of certain cultural markers.

The best way to study the subsistence techniques is through their material remains (animal bones, plant seeds and labour tools) and this has been performed in the cardial and epicardial Neolithic thanks to the relentless work of researchers such as Antolín (2013), Saña *et al.* (2015), Palomo (2012), Gibaja *et al.* (2018a) and many others (Ache 2011, Allué 2003b, Ballesteros 2009, Borrell and Molist 2012, Bofill *et al.* 2008, Buxó and Canal 2008, Bergadà 1998a and b, Colominas *et al.* 2008, Lloveras *et al.* 2014, Malgosa 2016, Mensua and Piqué 2008, Miró 1991, Morales *et al.* 2013, Nadal and Estrada 2008, Navarrete *et al.* 2017, Revelles *et al.* 2018, Spiteri *et al.* 2016). In this sense, cardial wares can also be studied as the result of a production process. Based on ethnographic studies (García-Roselló 2007) a "Chaine operatoire" belonging to a cultural group can be evaluated to detect possible shifts related to identity. Within this framework, studies on the raw materials indicated the tendency to use grog in cardial layers and calcite in more recent periods (Clop 2011, Binder *et al.* 2010a).

Therefore, the Cardial interpretative value as marker of the Neolithic package was built based on studies validating their existence across a time period measured with ^{14}C dates. Although not systematically, the development of bioarchaeology is now capable of detecting remains directly linked with the agricultural and herding practices in a certain period of the past. Nowadays, when finding a piece of Cardial pottery in a specific stratigraphic context it could be assumed that it belonged to the VIth millennia and was used by peasants and shepherds. Nevertheless, to prove it, ^{14}C, ancient DNA, plant and animal remains can be used more successfully, thus partially depriving pottery of its hitherto informative value.

1.2.3 Pottery, a canvas for decorative art.

The study of the cardial decorative technique in itself is not a forgotten line of research. The original description of the first motives used was performed by Colominas (1925) himself in the framework of an experimental process (creating impressed decorations with seashells on a piece of flat clay), which is at the core of the attribution of this decorative technique as the result of using a Cerastoderma edule as a tool. The description of the cardial tool was performed by different parameters (the active part of the tool, the position of the tool, the geometry of the decorative motive, the placement of the motive in the vessel and the gesture used (Figure 4). Nevertheless, this proposal was not fully systematised. Out of the 13 identified motives, none was described by the five presented variables. Therefore, the description was subjective. However, this method was coherent with the type of research conducted at that moment. Moreover, the types established by Colominas were then used by Grivé (1931) to describe the pieces from the Esquerda de les Roques del Pany. Therefore it should be stated that the theoretic base supporting this initial stylistic research was that different techniques and decorative motives could be used to indicate the identity of those who used it. Nevertheless, this premise was not applied to search identity differences between cardial motives, they were simply described in further publications. The data used to discuss the presence or absence of the cardial and epicardial cultures was focused in the combination of the different decorative techniques (impressions, incisions, plastic decorations).

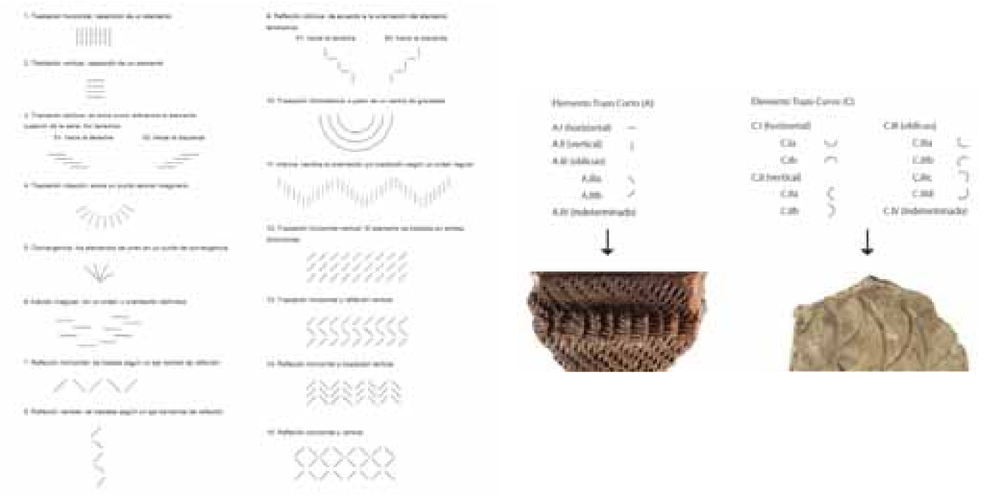


Figure 4: Codification system presented in Molina *et al.* 2010 and adapted for the systematic description of cardial decoration. Reproduced from Molina *et al.* 2010.

Finally, at the second half of the XXth century, new publications took the decorative motives back into account (Vilaseca 1945, Baldellou 1972, Baldellou 1974, Cura *et al.* 1979, Alcalde *et al.* 1991) or performed generic descriptions (cardial as a "baroque" decoration) (Martín *et al.* 1984, Gràcia 1987). Others just reported the presence of several decorative techniques (Virella 1973, Rovira 1976, Martin *et al.* 1981, Bosch and Aliaga 1998) with the single objective of establishing relative chronologies. The decorative motives did not had any interpretative value and research advanced (as previously stated) independently of the motives found in the published sites. Nevertheless, the efforts to describe them continued and, in the Monsterrat round table, descriptions of decorative motives were reported by Tarrús (1981) for the area of Girona and by Mestres (1981) for the site of

Guixeres de Vilobí. The used variables at this point in time were the essentially the same proposed by Colominas. This lack of advancement could be explained by the absence of the archaeological meaning the decorative motives had. The description was only performed as the process of presenting the assemblages to other researchers.

Nevertheless, Mestres (1981) tried to perform new interpretations which could have finally given decorative motives some interpretative value. It seemed that the biggest vessels would be associated with thicker and deeper impressions which smaller vessels presenting more soft and thin marks. This intuition was later pursued by studying the morphometry of the cardial imprint, which was directly linked to the dimensions of the shell used (Breu 2013).

Furthermore, Mestres also suggested that the vases which could have been older presented the most structured and complete decoration (filling all the surface of the upper part of the vessel). This proposal had certain continuity when Bosch *et al.* (2011) suggested the motive decorative would simplify over time or they would, as implied by Mannen (2002), vary.

Mannen's work (2002) was the first time differences on the decoration were used to suggest the presence of different styles which would have changed over time. The systematic and quantitative study of the data, thus, resulted in a synthetic approach which established which type of decoration could be expected from a certain place in time.

The approach combining decorative studies and absolute dating was further developed by Oms (2014) for the north-east of the Iberian Peninsula. After careful and exhaustive evaluation of the available data at that time, Oms was able to distinguish decorative changes throughout the second half of the VIth millennia cal BC and the first centuries of the Vth millenia cal BC. Whithin the parameters of the cardial technique, dragged gestures and the use of the shell natis tended to increase in frequency over time. Contrarily to the interpretations put forward by Mestres (1981) or Bosch *et al.* (2011), the complexity of the decoration also seemed to increase over time, a pattern which was accompanied by an increase in more elaborated and combinations of themes (Oms 2014). The study of the territorial variation of vessel decoration also constituted a novelty in the region. The distinction of discrepancies linked to specific geographical areas offered the opportunity to explore socioeconomic factors which may have fostered the existence of different identities within the common Cardial "koine".

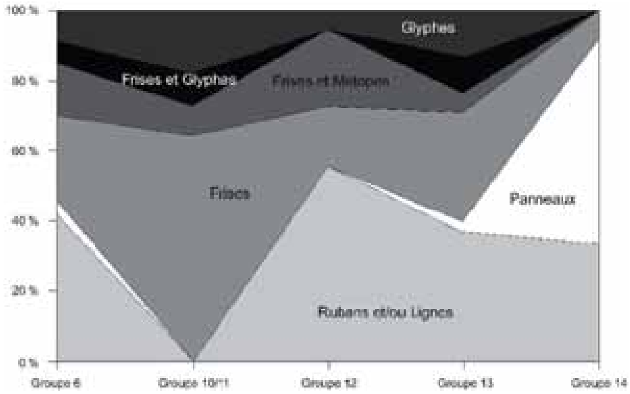


Figure 5: Percentage of decorative themes in different pottery types demonstrating the possible association between pottery shape and decoration. Reproduced from Molina *et al.* 2010.

The researchers in Valencia (Molina *et al.* 2010, Bernabeu *et al.* 2011), France (Manen 2002, Binder *et al.* 2010b) and Barcelona (Oms 2014) developed their own method for describing cardial decoration. They all aimed at detecting the grammatical rues and syntaxes governing the decoration. One of the most interesting lines of research would be linking the decorative motives and vessel functionality. With this idea in mind, Molina *et al.* (2010) evaluated several decorative motives in the central Mediterranean coast of the Iberian Peninsula. The 6 types of decoration detected (Glyphs, Friezes combined with Glyphs, Friezes combined with metopes, single Friezes, panels and lines) were compared with the ceramic shapes (bowls, jugs, pourers, jars, marmites and storage vessels) and it was noted that panel decorations were mainly found in storage vessels. This work therefore proved that such associations could be performed (see Figure 5).

In consequence, the arrival of interpretative proposals focused on the study of the decorative motives surpassed the idea of a homogenous cardial phenomenon. The differences in the decorative motives had been reported since 1925, but it is not until recently that said variations were significantly valued to understand the society of the VIth millennia cal BC.

1.2.4 Pottery at work, use and function.

When studying pottery as a tool, the archaeologist is equipped with two distinct but complementary strategies. The first is investigating the vessel's functionality. "Function" here is understood as the set of characteristics which make a tool better suited for a certain type of work. Within this framework, pottery shape, the selection of clays and surface treatments may be significant variables which can be taken into account (Clop 2002). The second is to approach pottery use. “Use” here is understood as the activities which were effectively performed by that artefact in the past. In this line of research, use-wear, inorganic/microscopic remains and organic residues may be key indicators. Therefore, both function and use link to different realities, one related with the design of the object itself and the physical limitations arising from it and another focused on the material consequences of the interaction of the artefact with other objects or products.

Regarding the study of pottery functionality within the Early Neolithic in the Iberian Peninsula, probably one of the most seminal and influential papers is the presentation of the typology of the vessels recovered until 1989 (Bernabeu 1989). The functionalistic nature of Bernabeu’s approach, rather than falling into risky overinterpretations, offered the opportunity to value the ceramic artefact as a tool within a certain margin of certainty. Some of the most interesting types highlighted by Bernabeu (1989) were necked vessels, whose shape would make them ideal for the storage, transport and drinking of liquids. Other really tiny small vessels were tentatively identified as pigment containers due to the presence of visible residues in the interior and its coincidence with the pigments in rock art. The existence of spouted vessels and significantly flat containers interpreted as dishes offered additional clues pointing at the possible uses different types of pottery might have had. The line developed by Bernabeu was followed by Bosch when studying the pottery from archaeological sites within the scope of this PhD. The studies from Plansallosa and La Draga (Bosch and Alliaga 1998, Bosch *et al.* 2000, Bosch and Tarrús 2018) highlighted that, when working mostly with rounded-based vessels, handles and appliqués would be necessary items to understand is possible handling strategies. Moreover, the dimensions inferred through the rim diameter and the wall width suggested the presence of vessels which could have been used to store liquids or solids (Bosch and Tarrús 2018). Studies from sites such as Cova del Vidre and Caserna de Sant Pau have also explored the mechanisms which could be in place to suspend essentially unstable pottery over the fire (Gómez *et al.* 2008, Oms 2014, Bosch 2016a).

Nevertheless, assemblages well preserved enough to perform a comprehensive study of vessel types are scarce in Early Neolithic period of the northeast of the Iberian Peninsula. When not possible, shapes have been approximated using other analytical terms such as “cylindric”, “hemispheric” or “subspheric” estimates and its combination with the artefact's dimensions and rim angles. In turn, these have been compared between assemblages to indirectly assess possible functional differences in different archaeological sites or contexts (Oms 2014). The interpretation of vessel types has also been helped by the nature of their archaeological context. For example, small vessels possibly for the preparation and consumption of food in the postcardial Neolithic burials in Caserna de Sant Pau may have played a different role than big containers, possibly for the transport and preservation of goods (Gómez *et al.* 2008). When comparing pottery typology to other variables, a range of publications have explored the shape as a marker for functionality and tried to assess whether a correlation with the decoration effectively existed (Flors i Sanfeliu 2011, Molina *et al.* 2010, Sanfeliu and Flors 2009, Binder 1991, Breu 2014). So far, it might seem that vessels potentially used for storage (higher dimensions) could have also presented simpler designs (Molina *et al.* 2010) in the Levantine coast of the Iberian Peninsula. The study of pottery motives from Grotte Lombarde in the south of France (Binder 1991) suggested that certain motives (ABC) could be associated to specific shapes (212) and that as the Neolithic advanced, shapes and decorations could have become more unstructured.

Objects connected to the pottery assemblages by stratigraphy might be of significance when exploring pottery function. In the Early Neolithic this would be the case of wood (Bosch *et al.* 1996), bone (Oms *et al.* 2016a, Bosch *et al.* 2016b) or even ceramic (Bernabeu 1989) spoons. The existence of these tools could be linked with stirring liquids and practices which might involve boiling, but direct evidences for these activities would be conditional to the performance of use-wear studies on said utensils.

Compared to functional studies, published data informing pottery use from the Neolithic in the whole Iberian Peninsula is still rather scarce. No known studies have reported use-wear patterns beyond possible soot from repeated exposure to fire (Berrocal 2018), which suggests that a comprehensive research on tribochemical wares on the oldest pottery from the Iberian Peninsula might be a possible line of research to improve the knowledge of the first farmers and shepherds. When looking at microscopic remains such as phytoliths which could constitute a residue of use, published data is extremely scarce. A Neolithic vessel from the Azután dolmen (Tresserras and Matamala 2005), several vessels from Ca l'Oliaire in the Middle Neolithic (4040—3680 calBC) (Martín *et al.* 2005) and one vessel from the Segudet tomb in Andorra (Yáñez *et al.* 2002) constitute some of the scarce examples of analyses reporting pollen, bacteria (diplococcus and streptococcus) and microalgae (diatoms) adhered to the inner pottery surfaces. Rather than interpreting these evidences on their own, researchers usually accompanied them with chromatographic studies of amorphous organic matter, putative organic residue analysis.

The analysis of hydrophobic matter via gas chromatography and mass spectrometry seems to have been the most successful approach to recover data relative to pottery use in the Neolithic of the Iberian Peninsula. Published studies of lipid analyses report that this technique has so far been attempted in roughly 120 pottery fragments from the whole region between roughly 5500 cal BC and 3000 cal BC. This includes studies from 10 sites: Cueva del Toro (Tarifa *et al.* 2019), Cova del Sardo (Tarifa 2015), Polideportivo de Martos (Sánchez *et al.* 1998), Cova de l'Or (Martí *et al.* 2009), Gavà (Tresserras 2009), Can Sadurní (Blasco *et al.* 2008, Spiteri *et al.* 2016), Segudet (Yáñez *et al.* 2002), Ca l'Oliaire (Martín *et al.* 2005) and the Azután Dolmen (Tresserras and Matamala 2005). Although these studies were successful in recovering significant amounts of interpretable organic matter only in half of the studied vessels, the obtained data was capable to support the existence of a wide range of products such as porcine and ruminant animal fats, dairy residues as well as plant waxes and resins. The encouraging results of this last analytical technique only stress the importance to reinforce future interdisciplinary projects which include use-wear and phytolith specialists to obtain the most complete view on pottery use possible. Regrettably, such a wide methodological approach would be unattainable in a single PhD. Therefore, when attempting to access pottery use, the technique of study of choice in this case has been organic residue analyses in hope that the possible relevance of the results will be able to motivate more comprehensive research projects integrating multiple lines of evidence.

Beyond organic residue analysis, pottery can be conceived as a surviving element of a complex system of interconnected social dynamics which may or may not be preserved in the archaeological record. In consequence, lithic tools, animal and human bones, charcoal, seeds, pollen, jewellery, hearths, pits, huts and many more must come together to inform the data obtained regarding pottery use and paint the most precise and complete picture possible. It is only by finding the material connexion in the archaeological register between the organic and the inorganic, the natural and the artificial, that pottery can be placed within past social frameworks and be judged for their actions.

The research on the artefact per se, which is most necessary and indispensable, will be complemented in this research with a focus on the elements which will link them with the other recovered pieces of evidence from the past. Notwithstanding, before discussing the archaeological questions which might be most informed by organic residue analyses, the potential, limitations and pitfalls of organic residue analyses must be laid out. To this end, the following pages will provide a concise review of the evolution of the discipline since its conception to the present day and the results which has been able to yield on pottery from the prehistory of the Iberian Peninsula.

1.3 The History of Organic Residue Analyses in the Iberian Peninsula and in the world.

1.3.1 The development of organic residue analysis.

The origin of residue analysis of archaeological vessels can be related to the appearance of the New Archaeology. In the late 1970s, when the academic environment was moving towards sciences, there was a strong interest to explore how the chemical characterisation of organic matter could provide new data from the past. Much of the necessary knowledge to perform organic residue analysis at that time was with geochemistry research teams, and, in fact, many of the first published papers analysed soils and sediments (Knights *et al.* 1983, Morgan *et al.* 1984). Nevertheless, it can be said that the 1970s and 1980s was a period of complete experimentation in which different researchers started to use molecular analytical chemistry to study ancient artefacts. Many archaeological materials such as mummies (Kuksis *et al.* 1978), coprolites (Lin *et al.* 1978), shipwrecks (Evershed *et al.* 1985), human tissues (Evershed and Connolly 1988), pottery (Condamin *et al.* 1976) and soils were analysed.

In the study of organic residues in pottery vessels, new research focused on providing results for case studies. The methodology was usually created ad hoc by adapting geochemical operating procedures while the identification of the compounds was performed by comparing the archaeological results with modern reference samples (Hurst *et al.* 1989). During that period, the main extraction procedure was based on the use of a Soxhlet apparatus and methyl ester derivatives from saponification (Condamin *et al.* 1976). These first identifications of olive oil, resins (Shackley 1982) or cocoa (Hurst *et al.* 1989) barely considered the impact that hydrolysis, oxidation and bacterial degradation could have on the chemical structure of the molecules. The interpretations of these researchers are nowadays subjected to criticism. Nevertheless, these pioneers made a first step by extracting meaningful quantities of lipids from archaeological vessels using both gas chromatography (GC) (Condamin *et al.* 1976) and high performance liquid chromatography (HPLC) (Hurst *et al.* 1989). All that remained was to develop a more accurate and systematic methodology that could provide more precise and realistic interpretations.

The 1990s witnessed many methodological advances. The use of Curie point pyrolysis was explored (Oudemans and Boon 1991) and the isotopic characterisation of visible residues (Hastorf and DeNiro 1985) would become a relevant approach (Sherriff *et al.* 1995). Nevertheless, the survival of food crusts has not been as widespread as the preservation of lipids embedded in the clay matrix of the vessel. Therefore, the paper that set the standard for the future of the study of embedded residues would be the publication of the chloroform/methanol and trimethylsilylation derivatisation extraction method in 1990 (Evershed *et al.* 1990). Many questions required an answer before residue analysis could provide reliable interpretations. Issues of contamination from soils (Heron *et al.* 1991), plastics and fingerprints (Evershed 1993) were studied. Thus realising that direct contact with human skin could transfer cholesterol and squalene to the sample and that molecules from the soil could also migrate inside the sherd during its burial.

Furthermore, one of the other main research directions was the assessment of the degradation processes that affected the residues. Hydrolysis of triacylglycerols, oxidation and biological degradation (through bacterial β-oxidation) were explored (Everhsed *et al.* 2002a) and the mechanisms explained. Later on, researchers like Regert *et al.* (1998) and Aillaud (2002) would further explore the by-products of the lipid degradation processes. Therefore, the publication of this research allowed molecules like dicarboxylic acids, hydroxy and dihydroxy acids, and ω-hydroxy acids to be incorporated into the interpretation toolset. The impact of microbial lipid contributions was also assessed at this time (Dudd *et al.* 1998) and suggested that, although biochemical degradation can be a major degradation factor, it takes a certain time for dead bacterial biomass to appear in the chromatograms. Nonetheless, the characteristic molecules are the same ones that can exist in ruminant fats, which forced the researchers to find other ways to detect these types of products.

One of the main advancements was the development of a theoretical background that situated the study of organic residues as one of the most powerful analyses of pottery use (Heron and Evershed 1993). The main questions of this research investigated the incorporation of residues in the clay matrix, and which areas of the vessel would be more likely to preserve higher quantities of lipids. Publications demonstrated that mixtures of residues were not uncommon (Charters *et al.* 1995), that vessel shape could indicate areas with higher lipid yield (such as rims or bases) (Charters *et al.* 1993b) and that cooking is one of the main causes of residue accumulation in vessel walls (Charters *et al.* 1997). The characterisation of lipids was led by a new way to interpret the residues, the biomarker concept. This approach is based on finding compounds and sets of molecules unique enough that only allow one conclusion. Therefore, the mere presence of C16:0 and C18:0 fatty acids in an amphora (Condamin *et al.* 1976) was not enough evidence anymore for the detection of olive oil. In several papers, experimental degradation tests and comparisons with modern day samples laid down a new set of biomarkers that would help the creation of a systematic method of identification. Specific molecules were proposed for the detection of animal fats (Evershed *et al.* 1990), leafy vegetables (Evershed *et al.* 1991), birch bark tar (Charters *et al.* 1993a) and beeswax (Heron *et al.* 1994). This set of molecules was enough for a start, however, as more samples were analysed, new biomarkers were incorporated.

One of the most interesting ones in terms of its interpretation power are long chain ketones. They are composed of any combination of C16:0, C18:0 and other free fatty acids and result from alteration due to exposure to temperatures higher than 300°C (Evershed *et al.* 1995a, Raven *et al.* 1997). Such a biomarker allowed interpretations about the actual social practices around pottery vases.

Nonetheless, one of the major advances in terms of biomarker development was the incorporation compound specific isotopic analyses through gas chromatography coupled to combustion and isotope ratio mass spectrometry (GC-C-IRMS) (Evershed *et al.* 1994). This analytical technique allowed acquiring isotopic ratios from specific compounds, which had the potential to improve residue interpretation (Evershed *et al.* 1997a, Evershed *et al.* 1997b). In its first stages, GC-C-IRMS data was used to develop new biomarkers and evaluate the degradation processes. Nevertheless, the significance of isotopic ratios significantly increased when the values were used to differentiate between animal fats. With the introduction of the dimethyl disulfide (DMDS) derivatisation (Francis

1981), the chromatographic separation of unsaturated fatty acids between double bond positions was achieved, which is a good biomarker for the differentiation of ruminant and non-ruminant animal fats (Mottram *et al.* 1999). Using this information, Evershed *et al.* (1997a) confirmed that δ^{13}C16:0 and δ^{13}C18:0 values were different between these two types of animals, thus providing an effective way to differentiate them.

Shortly after, isotopic ratio mass spectrometry was used to distinguish ruminant milk from ruminant adipose fats (Dudd and Evershed 1998), an approach that would be first tested with archaeological residues from the British Neolithic just one year later (Dudd *et al.* 1999). In consequence, by the year 2000 a wide range of biomarkers had been developed and residue analysis had achieved a mature state which made it a more realistic and accurate method. Evershed's (1999) review article was a way to sum up all the work done and, in a way, close a period of method development. Although new biomarkers would be incorporated in the future and the method would keep improving, the methodological foundations of organic residue analysis had been laid out and the technique was ready for use by archaeologists.

Meanwhile, researchers from other parts of the world were also starting to implement these new methods on the study of pottery use. Although countries like France or Germany had contributed to its development since the beginning (Condamin *et al.* 1976, Rottländer and Schlichtherle 1983), Spain only started to approach the relationship between organic chemistry and archaeology when ties had been sufficiently strengthened by international research. One of the early studies explored the potential of residue analysis in Roman and Medieval vessels (Cañabate and Sánchez 1995, 1996) while other researchers like Tresserras engaged with organic residue analysis as part of a multidisciplinary approach to residues in which phytoliths and starch also played a major role (Maya *et al.* 1999, Tresserras 1997a, Tresserras 1999, Yáñez *et al.* 2002). Nevertheless, papers were hard to access and the absence of any technical description of the procedures caused this early work to be completely inconspicuous beyond the borders of the most specialised Spanish academia. The analyses were usually performed on one or two samples, hence placing the results in a position in which they could not make any major contributions to the archaeological research. Therefore, although the method had been present amongst the Spanish academia, the common conception was that the technique was expensive, not reliable and, at times, completely irrelevant.

This latter opinion is the contrary of how residue analysis would be practiced in the XXIst century. Since it reached maturity, biomolecular archaeologists have pushed toward new areas of research by asking and answering relevant questions about how societies behaved in Prehistory. This research has focused on the actual contents of the pots and has opened a new window to understand the process of neolithisation and also the role of pottery amongst hunter-gatherers (Craig *et al.* 2013).

One example of this research is the question of the appearance of dairy products. After the development of a new biomarker for milk (Evershed *et al.* 2002b), residue analysis was in a position to test one of the main economic theories of prehistory, the secondary products revolution (Craig 2002). As it had been laid out by Sherratt (1981, 1997) herding management in the Neolithic would have been focused on meat production, whereas the arrival of the Chalcolithic and Bronze Age triggered the consumption of secondary products (wool and milk). For that theory to be correct, milk residues in pottery vessels should not be found in pots that predated the bell beaker horizon.

Nevertheless, a wide analysis of samples from Neolithic, Bronze and Iron Age Britain (Copley *et al.* 2003, Copley *et al.* 2005a, Copley *et al.* 2005b, Copley *et al.* 2005c, Copley *et al.* 2005d) demonstrated that dairying had been present as a part of the prehistoric economy since the Neolithic. Data from Scotland (Craig *et al.* 2005b), Romania and Hungary (Craig *et al.* 2005a), Switzerland (Spangenberg *et al.* 2006) and the Near East (Evershed *et al.* 2008b) also suggested that milk was a widespread product in the Neolithic and was present since the beginning of the use of pottery amongst agricultural and pastoralist societies. More recent research in other parts of Europe (Cramp *et al.* 2014a, Cramp *et al.* 2014b, Isaksson and Hallgren 2012, Salque *et al.* 2013) has placed Sherratt's secondary products revolution in a position much more difficult to defend, at least in terms of milk production. Although all research has focused on the Danubian branch of neolithisation, research on the Mediterranean region is testing the same hypothesis (Spiteri 2012, Spiteri *et al.* 2016). The prospects are that the milking of ruminant animals will be proven as an integral part of what it meant to be a Neolithic society.

Research on lactose intolerance carried out across Europe and the Near East has looked at DNA analysis from modern and ancient samples (Burger *et al.* 2007, Gamba *et al.* 2014, Ingram *et al.* 2009, and Lacan *et al.* 2011). Results

Residue	Example Origins	Main References
Animal Fats	United Kingdom, Kazakhstan, Italy, Spain	Mottram *et al.* 1999, Dudd *et al.* 1999, Mukherjee *et al.* 2008, Outram *et al.* 2011, Spiteri 2012.
Beeswax	Greece, United Kingdom, Jordan, Corsica	Heron *et al.* 1994, Evershed *et al.* 1997b, Evershed *et al.* 2003. Mazar *et al.* 2008, Namdar *et al.* 2009, Rageot *et al.* 2015.
Birch Bark Tar	United Kingdom, Greece, Corsica	Charters *et al.* 1993a, Mitkidou *et al.* 2008, Rageot *et al.* 2015
Bitumen	Middle East	Connan and Van de Velde 2010
Cocoa	Mexico	Hurst *et al.* 1989, Hurst *et al.* 2002, Washburn *et al.* 2014.
Dairy products	United Kingdom, Middle East, Romania, Hungary, Poland, Finland	Copley *et al.* 2003, Craig *et al.* 2005a, Spangenberg *et al.* 2006, Evershed *et al.* 2008b, Salque *et al.* 2013, Cramp *et al.* 2014a, 2014b.
Frankincense	Egypt, Belgium	Evershed *et al.* 1997c, Mathe *et al.* 2004, Baeten *et al.* 2014.
Lamp iluminants	Egypt	Copley *et al.* 2005e, Romanus *et al.* 2008.
Maize	United States, Argentina	Reber and Evershed 2004a and b, Lantos *et al.* 2015.
Marine prodcuts	South Africa, Denmark, Sweden, Lithuania	Copley *et al.* 2004, Craig *et al.* 2007, Craig *et al.* 2011, Isaksson and Hallgren 2012, Craig *et al.* 2013, Taché and Craig 2015, Heron *et al.* 2015.
Palm fruits	Egypt	Copley *et al.* 2001.
Plant residues	United Kingdom	Evershed *et al.* 1991, Colombini *et al.* 2005, Cramp *et al.* 2011.
Resins	Israel, United States, Turkey, Italy, United Kingdom	Fox *et al.* 1995, Regert *et al.* 2008, Reber and Hart 2008, Izzo *et al.* 2013, Brettell *et al.* 2014.
Wine	Egypt, Greece	Guasch-Jané *et al.* 2004, 2006a, 2006b, Barnard *et al.* 2011, Pecci *et al.* 2015, Garnier and Valamoti 2016

Table 1: Types of residues detected by organic residue analyses in pottery and the main papers supporting presenting their corresponding analytical data.

indicate that lactase persistence would have not been common through the Neolithic and, therefore, milk would have been an indigestible drink for any adult. Nevertheless, the transformation of this product to its dairy derivatives such as butter or cream would have removed the secondary effects of lactose. In consequence, the making of these products could have been part of the work load of any Neolithic society. This would be a dietary staple and a part of the everyday life that would have remained forgotten if it was not for the application organic residue analysis.

Another focus of research has been the question of pottery before agriculture. Many hunter gatherer societies in northern of Europe (Denmark and Finland as an example), Japan, and the north-eastern North America would have used pottery as part of their everyday life (Craig *et al.* 2013). Nevertheless, the actual use of these artefacts has remained a matter of speculation since highly mobile societies wouldn't depend on heavy and fragile objects like ceramic vessels. Research on detecting residues from aquatic resources was benefited by the detection of molecules such as ω-(o-alkylphenyl)alkanoic acids, polyunsaturated long chain fatty acids, isoprenoid acids (phytanic, pristanic and 4,8,12-TMTD acids) (Evershed *et al.* 2008a, Hansel *et al.* 2004) and a consistent enrichment of the $\delta^{13}C$ isotopic values of C18:0 and C16:0 fatty acids (Evershed *et al.* 2002b). Early studies showed fish products being used by farming and herding societies (Copley *et al.* 2004) but the study of hunter-gatherer pottery from northern Europe demonstrated a widespread use of pottery for the management of aquatic resources (Craig *et al.* 2007, Craig *et al.* 2011). These results have also been reproduced for north-eastern North America 1200 years before the arrival of maize (Taché and Craig 2015) and Jōmon pottery (Craig *et al.* 2013, Lucquin *et al.* 2016b).

The arrival of maize and its consumption has also been studied by residue analysis. The proposal of a biomarker (n-dotriacontanol) combined with compound specific isotopic values showing enriched values attributable to C4 plants were used in a comprehensive study of Missisipian pottery. Out of 134 sherds, 7 vessels containing maize oils were detected (Reber *et al.* 2004, Reber and Evershed 2004a, Reber and Evershed 2004b), which meant that maize was being distinguished in only 5% of the samples. More recently, Lantos *et al.* (2015) have proposed the use of the C16:0, C18:0 and C18:1 $\delta^{13}C$ isotopic values to detect fatty acids origination from C4 plants, which, when accompanied by the detection of starches in the same vessel, could suggest the presence of maize.

This new approach has been tested on samples from north-western Argentina and could suggest a second chance for the detection of the consumption of maize in America.

These are only a few examples of the potential of organic residue analysis when embedded in a wider archaeological context. Since the method reached its maturity in the beginning of the 21st century, many products have also been added to the interpretation toolset of organic residue analysis (see Table 1). Nevertheless, one country presents extensively more papers than the others, the United Kingdom (UK). This could come from the fact that UK was where organic residue analysis developed. On the other side of the spectra, countries like Spain seem not to be engaged in this type of analysis.

As previously presented, the state of residue analysis in Spain at the beginning of the 21st century was clearly not on par with international standards. Still nowadays there is a lot of grey literature being presented in small congresses and local journals (Guerra *et al.* 2012, Tresserras 2009). Nevertheless, new research exemplified by the work of Guasch-Jané on wine residues (Guasch-Jané *et al.* 2006a; 2006b, Guasch-Jané *et al.* 2004) has been able to enter the international scene.

Finally, a slow but steady change in approach seems to be broadening Spanish archaeologists' minds. The publication of articles in the local languages from international researchers (Craig 2006) and the impact that research on milk has had worldwide proved that, far from being expensive and irrelevant, residue analysis could make a difference. In this context, new students have undertaken PhD research with the aim to reach the international quality and to implement the most recent methodologies to the analysis of the prehistory of the Iberian Peninsula (Oltra 2010, García 2013, Genera *et al.* 2015, Molina 2011, Tarifa 2015). The work of Molina (2015b) on Argaric vessels and the research from University of Jaén, shows that the Spanish academia is once again engaged with this type of analysis (Manzano *et al.* 2015, Sánchez *et al.* 2011).

This change in approach is a clear example of the main challenges that residue analysis faces. The accomplishments of this technique have attracted researchers from all over the world, whom expect their potsherds to be analysed. Although the technique's basic principles were universal, preservation ratios in environmental regions other than the northern European climates were affected by harsher degradation conditions. In the Near East, lipid extraction using dichloromethane and methanol (Evershed *et al.* 1990) was only able to produce positive results in around 10% of the samples (Evershed *et al.* 2008b, Gregg and Slater 2010). The use of acid-base (Regert *et al.* 1998) and microwave assisted extractions (Gregg and Slater 2010) were the first attempts to solve the problem of differential geographical preservation, and allowed yields to be significantly raised. More recently, Correa-Ascencio and Evershed (2014) published a new extraction method based on the use of acidified methanol. This new approach has also dramatically increased the number of positive samples, though its applicability might not be optimal for environments with good preservation. It is clear that, as time passes, not only extractions but also the understanding of degradative processes and isotopic biomarkers will have to be adapted to the different environmental conditions in which the potsherds were buried for thousands of years.

1.3.2 Organic Residue Analysis in the Iberian Peninsula, trends and future prospects

Recovering data regarding the ancient contents of an archaeological vessel is a great asset in developing well-founded interpretations of how past societies developed. Detection of animal fat, oils, milk, dairy products, fish fat and even wine have provided an important inside towards rituals, exchange and production systems. As previously reviewed, since the late 1970', chemists and archaeologists have collaborated in finding the trace lipid molecules indicative of different pottery contents. To contribute to its progress, the achievements of lipid organic residue analysis in the Iberian Peninsula and the limitations that the present sample preparation and analytical techniques show must be evaluated first.

It is not the aim of this chapter to present an extended literature review of the state of organic residue analysis in the Iberian Peninsula. The recent work by Guerra (2006) and Molina (2015b) present a wide coverage of the studies published both internationally and locally. Nowadays, the archaeological value of lipid organic residue analysis is the acquisition of additional data on the possible contents the vessels had. Sometimes interpretations transcend the pure description by using the data to assign different uses to the spaces where the pottery was found (Sánchez *et al.* 1998). Residue characterisation has been used to discuss the symbolic and ritual roles bell beaker pottery could have had (Guerra 2006). Nevertheless, the general lack of a clear archaeological question solvable through these analyses has relegated reports on pottery residues to annexes (Clausell *et al.* 2000), footnotes (Castro *et al.* 2012) and purely descriptive reports (Tresserras 2009), a place where they hardly help make an impact into the current knowledge and understanding of prehistoric societies.

Organic residue analysis offers the chance to tackle the active role of pottery in society. Usually, this line of research is performed by assessing the correlation between vessel shape and content (Sánchez *et al.* 2011: 224). Another option is to detect the presence of certain products which would be otherwise undetectable and thus prove the exploitation of certain natural resources and its specific production processes. Milk, honey and fish are just some of these products. As discussed, previous research has answered whether deep dietetic changes in the Mesolithic-Neolithic transition in northern Europe existed (Cramp *et al.* 2014b), whether consumption of dairy products was widespread despite of lactose intolerance (Evershed *et al.* 2008b) or whether a close relationship existed between fish products and pottery in hunter-gatherer societies (Craig *et al.* 2013). Nevertheless, although answers to these questions are available for many other European regions, organic residue analyses in the Iberian Peninsula have not been able to provide enough quality data to clearly assess these questions.

Therefore, there is a need to understand the way organic residue analysis is performed in the Iberian Peninsula in order to find the best strategies capable of yielding the best quality data with the most possibilities for archaeological interpretation. A wide (but not complete) literature review of 38 reports of

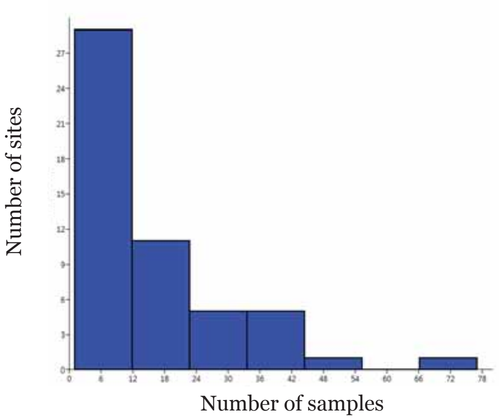


Figure 6: Histogram presenting the number of pottery samples picked from sites where analyses have been performed.

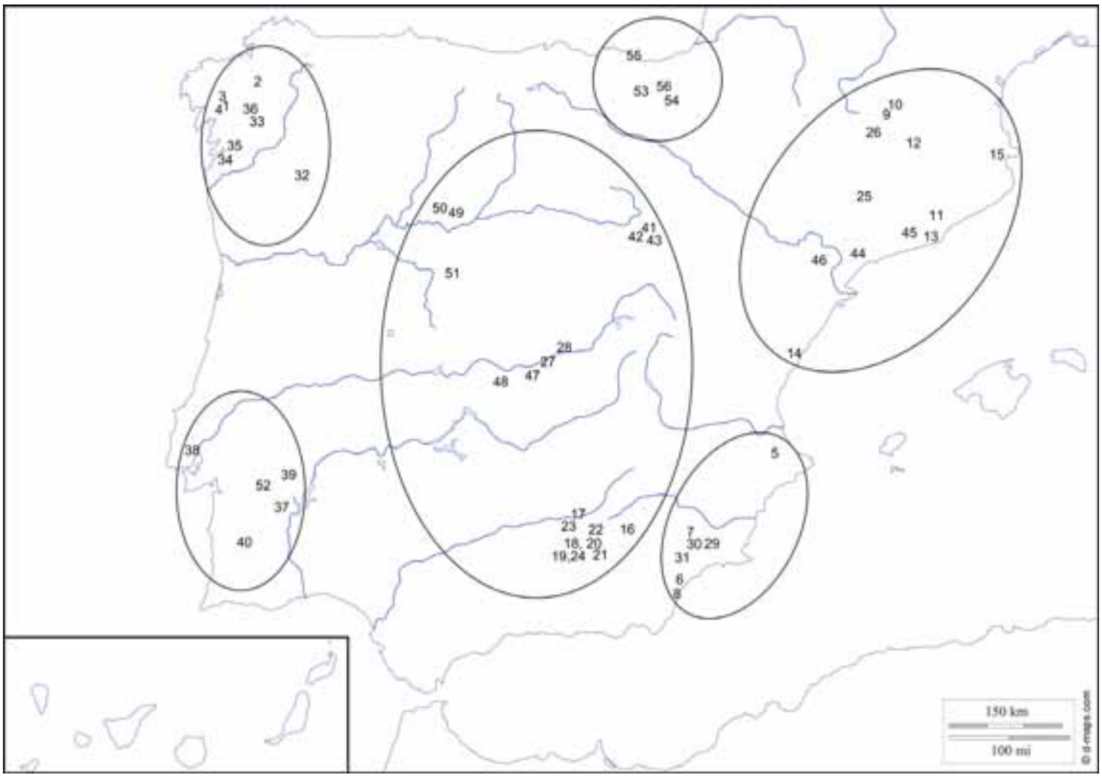


Figure 7: Approximate location of the archaeological sites reported in the literature with potsherds analysed by GC-MS in order to detect embedded lipid residues. 1 Devesa do Rei (Prieto *et al.* 2009), 2 Forno dos Mouros (Prieto *et al.* 2005), 3 Mirás (Prieto *et al.* 2005), 4 Agronovo (Prieto *et al.* 2009), 5 Cova de l'Or (Martí *et al.* 2009), 6 Fuente Álamo (Tresserras 2004), 7 La Almoloya (Molina, 2015b), 8 Gatas (Oltra, 2010), 9 Fossa de Prats (Yáñez *et al.* 2002a), 10 Tomba de Segudet (Yáñez *et al.* 2002b), 11 Can Roqueta (Tresserras 1997b), 12 Ca l'Oliaire (Martín *et al.* 2005), 13 Gavà (Tresserras 2009), 14 Torrelló del Boverot (Clausell *et al.* 2000), 15 Mas Castellar (Tresserras 2000a), 16 Castellon Alto (Parras *et al.* 2011), 17 Peñalosa (Manzano *et al.* 2015), 18 Remojadero de Pescado (Sánchez and Cañabate, 1998), 19 Polideportivo de Martos (Sánchez *et al.* 1998), 20 Marroquíes Bajos (Cañabate and Sánchez, 1997), 21 Cerro el Pajarillo (Parras *et al.* 2015), 22 Puente Tablas (Parras *et al.* 2015), 23 Las Atalayuelas (Sánchez *et al.* 2011), 24 Puente la Olla (Cañabate and Sánchez 1995), 25 Genó (Maya *et al.* 1999), 26 Cova del Sardo (Tarifa, 2015), 27 Humanejos (Rios *et al.* 2011), 28 Camino de las Yeseras (Rios *et al.* 2011) 29 Tira del Lienzo (Molina, 2015b), 30 La Bastida (Molina, 2015b), 31 Murviedro (Oltra, 2010), 32 A Froxa (Prieto *et al.* 2005), 33 Caballeira do Espíritu Santo (Prieto *et al.* 2009), 34 San Cosme 3 (Prieto *et al.* 2005), 35 Monte Buxel (Prieto *et al.* 2005), 36 Monte de Os Escurros (Prieto *et al.* 2009), 37 Castro do Ratinhos (García, 2010), 38 Bela Vista (Bastos, 2013), 39 Perdigoes (Bastos, 2013), 40 Garvao (Rosada *et al.* 2014), 41 Valdepernales (Rojo-Guerra *et al.* 2008), 42 Abrigo de Carlos Alvarez (Rojo-Guerra *et al.* 2008), 43 Los Dolientes (Rojo-Guerra *et al.* 2008), 44 Puig Roig del Roget (Genera *et al.* 2015), 45 Can Sadurní (Spiteri, 2012), 46 Tosal Montañés (Tresserras, 2006), 47 Valle de las Higueras (Bueno *et al.* 2005), 48 Dolmen de Azután (Tresserras and Matamala, 2005), 49 San Bernardo (Guerra *et al.* 2012), 50 El Nogalillo (Guerra *et al.* 2012), 51 Almenara de Adaja (Guerra, 2006), 52 Valada do Mato (Salque *et al.* 2015), 53 Mendandia (Salque *et al.* 2015), 54 Los Cascajos (Salque *et al.* 2015), 55 Kobaederra (Salque *et al.* 2015), 56 Atxoste (Salque *et al.* 2015) 57 Cueva del Toro (Tarifa *et al.* 2019) 58 Cueva Virués-Martínez (Manzano *et al.* 2019) source: self elaborated.

organic residue analysis performed in the Iberian Peninsula revealed that, up to date, more than 700 vessels dating between the first Neolithic and the Roman conquest have been analysed from 52 sites (Figure 6 and 7). Nevertheless, when critically assessing these reports two main tendencies seem to appear:

1 Small sample sizes: From the 52 sites with studies that report the number of analysed samples, barely 20% present sample sizes higher than 30 (see Figure 6). In fact, only 50% of the studies report more than 10 samples. The extent of this problem has to be understood in a research field, archaeology, in which the access to remains from the past and the funds are neither easy nor unlimited. Nevertheless, in the case of pottery and potsherds, which are usually the most common archaeological remains, these sampling problems could be easily solved.

2 Preservation rates: It is a common practice amongst international publications to report the amount of samples that yielded above the $5g{\cdot}g^{-1}$ lipid threshold for significant presents of fats. The rate between all the analysed samples and those with a positive result is called the preservation rate and aims to provide an estimate for the number of samples that might not yield any residue. This problem rises from the actual impossibility to detect whether lipids survived in the potsherd without performing the analysis, which is completely destructive. Nevertheless, when assessing the extent of post-depositional degradation processes, the possibility that the vessels never interacted with any fatty acids must also be taken into account. Therefore, low preservation rates might be explained by two possibilities: a) the environment in which pots were buried highly damaged the lipids embedded in the clay matrix, b) only a reduced amount of vessels ever contained fats. The only way to clearly assess one of these factors is by knowing the extent of the other. In the case of the Iberian Peninsula, its geography creates significant climate differences between the seashore and inland. In order to take these possible variations into account, (Figure 7) sites have been grouped in regional clusters based on shared broad climatic conditions. Two regions (the North West and the Basque County) could not be evaluated due to the number of positive results not being fully reported in the studies. Also, preservation rates were not obtained in sites with less than three studied samples. The other regions are presented in Table 2. In order to account for possible wide general trends in the consumption of more or less fatty products, preservation rates are also reported separated by the 4 main prehistoric periods.

		Number of shreds reported	Reported positive results	Mean preservation rate (%)	Number of sites
Regions	North East	175	51	0.29	10
	South East	257	120	0.41	7
	Centre	136	29	0.21	10
	Athlantic	47	15	0.32	2
Chronology	Neolithic	115	57	0.49	6
	Chalcolithic	104	35	0.33	8
	Bronze Age	267	105	0.39	12
	Iron Age	119	20	0.17	6
Sites with more than 3 samples		605	217	0.36	32
Sites with at least 1 sample		638	250	0.39	45

Table 2: Preservation rates for each region and chronology in the Iberian Peninsula. Sites with just one sample analysed are included to show their little effect in the overall preservation rate.

As it can be seen, regardless of prehistoric period or place in the Iberian Peninsula, preservation rates hardly exceed 50%. Moreover, only in a minority of sites (10%) (Martí *et al.* 2009; Molina 2015b, Manzano *et al.* 2015) the number of positive results is higher than the number of potsherds that yielded virtually no lipids. A comparison between these values and reports from other European regions (Table 3) clearly suggests that the climate plays a key role in explaining why fewer percentages of pots present interpretable lipids.

Region	Preservation rate range	References
British Isles	50 to 60%	(Mukherjee *et al.* 2005; Evershed *et al.* 1997a).
Eastern Mediterranean	20 to 30%	(Thissen *et al.* 2010; Evershed *et al.* 2008b).
Italic Peninsula	12 to 24%	(Salque *et al.* 2012; Spiteri, 2012).
Iberian Peninsula	17 to 49%	Table 2.

Table 3: Comparison of the Iberian preservation rate with other European regions.

Nevertheless, a third factor might be into play in order to explain preservation rates, the process of lipid extraction itself. Assuming no chemical procedure will perfectly extract all lipids, a choice must be made between strong base extractions (Regert *et al.* 1998, Evershed 2008, Correa-Ascencio and Evershed 2014), designed to fully recover all organic matter, specially the lipids more tightly bound to the ceramic matrix (Correa-Ascencio and Evershed 2014), but partially degrading its chemical structures, or “soft” extractions (Evershed *et al.* 1990), which will preserve the integrity of the molecules but create false negatives (samples that may contain significant lipids are not detected). Therefore, where climatic conditions have allowed good lipid preservation, strong base extractions would reduce the interpretation potential while producing the same amount of false negatives. Nevertheless in highly hydrolysing and oxidising environments where fat would have already degraded, strong extractions would not deteriorate organic matter further (thus interpretation potential would remain the same) but significantly reduce the amount of false negatives (Correa-Ascencio and Evershed 2014).

When reported in the literature, two main extraction procedures seem to be used in the Iberian Peninsula. The first is the classical Chloroform/methanol "soft" extraction (Evershed *et al.* 1990). It is the most widespread procedure applied for lipid extraction from potsherds (ex. Copley *et al.* 2005a; Evershed *et al.* 2008b) and, therefore, it has also been widely used in the Iberian Peninsula (ex. Oltra 2010, Parras *et al.* 2011, Bastos 2013, Genera *et al.* 2015, Tarifa *et al.* 2019). The second more extensively used procedure is the extraction with a Soxhlet apparatus and a range of organic solvents (Tresserras 1997a, Tresserras 2004, Tresserras 2006). Nevertheless, this last extraction is hardly acknowledged by their users (ex. Clausell *et al.* 2000, Tresserras 1999), making it difficult to evaluate the extractive capacity of their technique, although it would be safe to assume that only "soft" extractions were performed. In consequence, it might seem that a technique designed for environments with good organic matter preservation and weak lipid bonding has been repeatedly used for the Iberian Peninsula. In light of this fact, it should be evaluated whether the decrease of preservation rates for Mediterranean environments (Table 3) is partially caused by reported false negatives.

Once lipids have been extracted and analysed, three types of residues are commonly detected in the Iberian Peninsula: Beeswax (Heron *et al.* 1994), degraded animal fat (Regert *et al.* 1998) and well preserved fats. Plant oils and other highly interesting indicators such as thermal degradation (Raven *et al.* 1997, Evershed *et al.* 1995a, Hansel *et al.* 2004) are very scarcely reported.

The interpretation of lipid profiles is not straightforward. Because analytical results yield data that is not directly usable by the archaeologist, lipid profiles must be translated into specific products such as animal fat, milk or honey. The method used for this translation is the biomarker concept. Following this principle, organic products can be defined by a group and quantity of molecules which, when detected in the chromatograms, will prove presence of that product. Archaeological biomarkers for a wide range of products have been developed taking degradation effects such as hydrolysis and oxidation into account (Dunne *et al.* 2012: 342). As previously mentioned, a key biomarker for detection of milk, differentiation between different types of animal fat and marine or terrestrial products is the $\delta^{13}C$ value for Plamitic (C16:0) and Stearic (C18:0) fatty acids. These values are obtained through GC-C-IRMS (gas chromatography coupled to combustion and isotope ratio mass spectrometry), which is complementary to the common GC-MS analysis and only performed when molecules in the sample have been correctly identified and separated. So far, in the Iberian Peninsula, reports of GC-C-IRMS analysis of stearic and palmitic acids is scarce and only reported for some Neolithic and Bronze Age sites (Molina 2015b, Martí *et al.* 2009, Oltra 2010, Manzano *et al.* 2015, Breu 2015, Spiteri *et al.* 2016, Tarifa *et al.* 2019). Nevertheless, the interpretation of its values, although it holds a lot of potential for the understanding of prehistoric societies, presents some problems. Reported Iberian samples present a clear offset towards more enriched isotopic values. Experimental analyses performed at the island of Malta (Spiteri 2012) provided values for animal and milk fatty acids that support this enrichment, which had already been detected for other world regions (Mukherjee *et al.* 2005).

Therefore, it seems clear that a shift possibly due to climatic conditions exist in the carbon isotopic values of organic molecules. These changes are rooted in the way plants, the base of the terrestrial food chain, incorporate carbon and engage in diverse fractionation processes. Many factors might modify the $\delta^{13}C$ values of plant tissues. Light, nutrients, temperature and water availability would make the values increase. Nevertheless, deciduous trees, woody plants having higher $\delta^{13}C$ values tan herbaceous and altitude might suggest values would also descend. Finally, as climate changes through the territory new more adapted species tend to appear (Heaton 1999). These will present lower $\delta^{13}C$ values than its “less adapted” counterparts. Thus values seem to differ up to 2‰ depending on the region (van Klinken *et al.* 2000). If isotopic values from organic residues are interpreted without taking this possible shift into account, there is the risk of performing incorrect product assignations, thus affecting its archaeological significance.

1.3.3 Rethinking the sample preparation and interpretation.

Taking all the limitations of the method laid out previously it is clear that different preservation environments need different analytical approaches. Following this idea, three changes on the way lipid organic residue analysis is performed in the Iberian Peninsula could highly improve its applicability to archaeology:

- Sampling strategies: A careful evaluation of the archaeological contexts will help target the vessels with the highest historic significance. This must be complemented by the goal to sample the highest possible number of vessels so that statistically meaningful studies can be performed. Nevertheless, given that the nature of the analysis is destructive a comprehensive understanding of the potsherds will be of vital importance not to unintendedly duplicate analyses. Albeit achieving a large sample size might not be possible in all archaeological sites, the limiting factors should be made available.

- Switch to strong base extractions. New extraction procedures such as the acidified methanol extraction could be able to reduce false negatives (Correa-Ascencio and Evershed, 2014) and also lower amount of sample needed for analysis (Papakosta *et al.* 2015). Given that this is still a young procedure it is recommended to evaluate the extent of degradation with DCM/MeOH extraction first (Tarifa *et al.* 2019). The acidified methanol extraction has already been used in context from other world regions with highly promising results in terms of preservation rates (Taché and Craig 2015).

- Although palaeodietary studies on bone collagen will need specific reference samples for the correct interpretation of the isotopic values, in the case or organic residues, milk can still be distinguished from adipose fats because the different biochemical processes in the animals digestive system act on top of the already enriched plant values. Mukherjee *et al.* (2005) suggest the use of the $\Delta^{13}C$ values ($\delta^{13}C18{:}0$ – $\delta^{13}C16{:}0$), which correct for almost all of the environmental effects, thus allowing a correct interpretation of the results. Nevertheless, although this approach partially resolves the climatic issues, it also removes other variation factors which could be of archaeological interest; one of these is the possible mixture between different species of animal fats. When mixture of different types of species is suspected, the only way to assess it is to revert to the specific molecular values and apply mathematical mixing models (Mukherjee *et al.* 2005, Fernandes *et al.* 2017), nevertheless, this exercise will need new a careful evaluation of the available reference fats for animals and plants raised in different climates.

In conclusion, rethinking the way lipid organic residue analysis is performed in the Iberian Peninsula might open the possibility to ask new archaeological questions that so far might seem difficult to answer. Was dairying a widespread practice with the arrival of the Neolithic? What was the extent of fish consumption by social groups located in the seashore? It is clear that, by implementing more optimised methods, new possibilities are open for discussion.

2 Aims, objectives and rationale.

To build a discourse capable of explaining changes in the past is one of the main goals of archaeologists and historians. Nevertheless, as it happens with many other disciplines incorporating the time dimension such as astronomy, geology, and paleoclimatology, data are usually limited, fragmented and biased by a wide range of factors. Therefore, using all available information from several independent lines of evidence becomes a necessity to gain new insights on temporal changes. The aim of this PhD research is the study of Early Neolithic diets and culinary practices by exploring several historic questions related with the spread of the Neolithic way of life (agriculture, pastoralism and beyond). This will be achieved through the analysis of the organic matter preserved in pottery vessels from the north-east of the Iberian Peninsula in a highly significant moment in history when this artefact-type was first introduced. Organic residues informing about pottery use provide a unique opportunity to link multiple lines of evidence characterising the first societies such as animal bones, charcoal remains, plant seeds, use-wear patterns from lithic tools and the shape and characteristics of pottery itself. To this end, two general objectives have been devised:

1 Evaluate the relevance of plant, terrestrial and marine animal products in the Early Neolithic diet in the north-east of the Iberian Peninsula.

2 Understand how culinary practices may have varied in the period and region under study.

To fulfil these, six more specific objectives have been devised:

-Assess the existence of dairy fats and their possible link with herding strategies.

-Evaluate differences in the culinary practices through different settlement types.

-Explore regional variations in pottery contents across the territory.

-Detect differences in pottery contents between the coast and inland.

-Examine changes in culinary practices between the Cardial and Epicardial periods.

-Investigate associations between organic residues and pottery types.

The rationale behind these goals is based on relevant archaeological questions affecting the Early Neolithic diets and culinary practices across Europe. Therefore, its careful evaluation in a constrained and well-defined region, the north-east of the Iberian Peninsula, offers an opportunity to better inform lines of research active across the continent from a regional perspective. At the same time, it will contribute to the improvement of the knowledge available on the first farmers and herders in these territories from a novel perspective, organic residue analysis.

2.1 Research questions on the Early Neolithic diet: dairying in the Mediterranean neolithisation route.

Adult hypolactasia, the reduction of the production of lactase after childhood, is a common trait of most mammals (Leonardi 2014). However, lactase persistence in adults is an unevenly distributed phenomenon unique to the Homo sapiens. Genetic studies indicate that it is the result of convergent evolution dominated by the -13.919:C<T allele (combined with -22.018:A) in Eurasia, North Africa and Central Africa and four other mutations (13.915:G associated with the Middle East and 13907:G, -13.009:G and -14.010:C associated with East Africa) (Tishkoff *et al.* 2007). The appearance of lactase persistence dated through molecular clocks places these genotypes around the time of widespread animal domestication in the old continent (ex: Itan *et al.* 2009, Bersaglieri *et al.* 2004), albeit with high margins of error. When approximate estimates for the population at that moment in time are taken into account, the resulting selection coefficients suggests that the spread of lactase persistence is amongst the most strongly positively selected traits in human genetic history (Ségurel and Bon 2017).

Although local explanations including the calcium assimilation hypothesis (vitamin D deficiency) for high north latitudes (Flatz and Rotthauwe 1973) and the dry arid environment hypothesis for some places in the Middle East (Cook *et al.* 1978) provide plausible causes for specific cases, the western Mediterranean would not be coherent with any of them. Alternatively, the cultural-historical hypothesis assumed that those groups more reliant on animal products would have therefore suffered from higher selection pressures to become lactase persistent. In this case, societies with a high pastoralist heritage should be found to present higher ratios of lactase persistence. Although, to a degree, this seemed to be the case of Africa, an ethnographic study of 28 Eurasian populations did not find any correlation between the intensity of pastoralism and the frequency of lactase persistence (Holden and Mace 1997). Nevertheless, testing this historical hypothesis via the single use of modern populations poses several limitations related to genetic mixing and the assumption that a certain genetic group would have been consistently linked with a specific economic strategy (such as pastoralism) for millennia. Moreover, it has been widely discussed whether the dietary constraints imposed by the negative symptoms of lactose intolerance might have been tamed by specific non-genetic traits (Holden and Mace 1997). An adapted colon microbiota and the preparation of dairy products via fermentation, lactase autodigestion or the separation of lactase via curdling might reduce the negative effects of their malabsorption (Bayless *et al.* 2017), which does not support a situation which would result in significantly high selection coefficients.

So far, ancient DNA analyses have not been successful in detecting the -13.910:C>T mutation before 5000 BP (ex: Lacan 2011, Sverrisdóttir *et al.* 2014, Plantinga 2012, Burger *et al.* 2007, Olalde *et al.* 2018), which suggests that, if present, lactase persistence would have been extremely rare in the Neolithic. Nevertheless, the management of dairy products has been directly linked with the exploitation systems in place at the moment of animal domestication via organic residue analyses and its correlation with cattle herding in the Middle east (Evershed *et al.* 2008b). This suggests that pastoralism might have still played a significant role into the appearance of lactase persistence. Although genetic pressure to cattle could be assumed for the Danube neolithisation route (Beja-Pereira *et al.* 2003), the situation is less clear in the Mediterranean. The structure of the herds in southern Europe presented significantly higher amounts of sheeps and goats (Saña 2013, Manning *et al.* 2013a, Manning *et al.* 2013b) and the incidence of dairy products in pottery vessels has been shown to significantly vary across the mediterranean. While these ceramics are scarce in the Fertile Crescent, north-western Anatolia concentrates a significantly high amount of residues associated with milk fats (Evershed *et al.* 2008b). Evidences vary again across the Aegean, Adriatic and Tyrrenian (ex: Spiteri 2012, Salque *et al.* 2012) but there is no sufficient data available from the Iberian Peninsula to clearly assess their incidence. This might be because direct detection of dairy products within pottery vessels is still constrained to a handful of sites across central and western Mediterranean (Spiteri *et al.* 2016, Salque *et al.* 2012). Furthermore, a recent review by Spiteri *et al.* (2016) set the debate on dairy production strategies in the Mediterranean neolithisation route. In this paper, Spiteri highlighted the possible relevance of sheeps and goats as milk producers and pointed at the possible disparity in dairy products between caves and open air sites due to differences in the ruminant exploitation system.

In consequence, further assessment of the dairying practices in the Mediterranean is necessary to obtain a complete picture informing about the Neolithic social dynamics which could have resulted in the strong selection of lactase persistence. Recovering data informing about dairy production strategies is therefore linked to improving our understanding of the range of genetic traits selected at the moment of domestication (DNA changes in animals) and, therefore, the origins of the Neolithic phenomenon itself.

To fulfil this goal, it will be necessary to perform a comprehensive sampling of Early Neolithic pottery from western Mediterranean caves and open air sites and a different range of herd compositions in order to detect evidences of dairy products.

2.2 Questions regarding Early Neolithic diets: the relationship between the first farmers and the sea.

Across the Mediterranean, the study of fish and mollusc remains as well as human collagen isotopes has been consistently trying to assess the nature of the management strategies in place around marine resources. Solving this question is a key piece of the puzzle when the spread of the Neolithic across the Mediterranean is studied. As discussed by Rainsford (2014), the implications of fishing go beyond the yes/no or wild/domestic dichotomy as the consumption and exploitation patterns would be inserted in the social practices of the period. Cardial decoration evidences the use of seashells and several pieces of jewellery indicate the existence of marine products within the most symbolic spheres of society. Therefore, when trying to understand the role of the marine products, all lines of evidence become necessary.

As suggested by Desse (1987), fishing in the mediterranean Neolithic could be studied thanks to the improving excavation techniques but, at that time, they seemed to show rather local fishing strategies with positive evidence of fish consumption such as traces of burning in the bones. This picture has been expanded and complemented over time thanks to other archaeological records such as Franchthi Cave (Mylona 2014), Youra (Powell 2003) and Vela Spila (Rainsford 2014). Overall, most authors seem to agree on the existence of a small-scale coastal opportunistic fishing which would mark the decrease in aquatic resources and the intensity of their exploitation with the arrival of agriculture and pastoralism (Vika and Theodoropoulou 2019, Powell 2003, Raisnford 2014, Lightfoot *et al.* 2011). But this was not always the case, more than 50% of the bone assemblage in Grotta dell'Uzzo belonged to fish (Tagliacozzo 1994) and more than 2000 marine remains were recovered from the Early Neolithic layers in Cova de les Cendres (Marlasca 2013). Other sites in the western Mediterranean present further evidence for an increased fishing activity. In cueva de Nerja, although the number Early Neolithic fish bones was 53, the relative quantity of marine remains underwent a significant increase when compared with the epipaleolithic layers (NR=8)(Aura *et al.* 2001) and, on the Atlantic side, El Retamar (Cádiz) provides further evidence for the reliance on marine products (Soriguer *et al.* 2002). Furthermore, although important quantities of fish remains are not widespread within the Iberian Mediterranean seashore, most of the coastal sites have significant seashell assemblages. This suggests the possible existence of a different model where the first agrarian explorations had to be supported with other sources of food, maybe from a marine origin (Marlasca 2013), thus resulting in the increase in consumption of such foods compared with their Mesolithic counterparts.

Isotopic studies on human bone collagen offer complementary evidence limited by the explanatory power of $\delta^{13}C$ and $\delta^{15}N$ values. It seems to be widely accepted that, because the studies detect a mean value accounting for the overall diet, aquatic inputs of less than 20% would be extremely difficult to differentiate from a terrestrial based diet (Milner *et al.* 2004, Richards and Schulting 2006). Moreover, it has become increasingly challenging to evaluate the impact of marine resources within Early Neolithic diets in the western Mediterranean due to a lack of human burials which can provide sufficient data (Salazar-García *et al.* 2018). Alternatively, data from hunter-gatherers and middle-late Neolithic burials can be used to complement the scarcity of studies in the VIth and early Vth millennia. Nevertheless, the most recent analyses suggest that, rather than a dietary change, the Mediterranean exploitation of marine resources would have continued the pattern already in place by the previous-hunter gatherers (Salazar-García *et al.* 2014b, Salazar-García *et al.* 2018), which was marked by a terrestrial diet containing plants using the C3 carbon fixation pathway.

Therefore, organic residue analyses on pottery from the Early Neolithic may provide a new line of evidence to inform this discussion. Even in cases with extensively vessel reuse involving terrestrial products, the detection of marine biomarkers would help assess their relevance in the Early Neolithic diet. The ω-(o-alkylphenyl) alkanoic acids of 20 and 22 chain lengths and 4,8,12-TMTD acids are molecules commonly detected in pottery organic residue analyses and specific for marine products (Cramp and Evershed 2014). Exploring the organic matter preserved in pottery is of significant importance to access data regarding the techniques involved in preparing marine products for consumption. Rather than focusing on sites with significant fish bone assemblages, it is also necessary to assess the significance of marine products within cases where the main aquatic resources recovered were marine seashells. This strategy might help to obtain a more complete picture which can inform the fishing increase, the fishing decrease or the fishing continuity hypotheses.

To fulfil this goal it will be equally necessary to perform a comprehensive sampling of Early Neolithic pottery from western Mediterranean sites with and without evidences for marine resource exploitation in order to assess the presence of marine biomarkers within its residues.

2.3 Questions regarding Early Neolithic diets: plant matter and non-culinary uses of pottery.

A review of the contents of organic matter within ceramic vessels in the Mediterranean region indicates that preservation is significantly lower than in artefacts from higher latitudes. The consequence of this phenomenon is that, for similar extraction mechanisms, significantly higher amounts of destructive analyses have to be performed to achieve comparable results. As detected by Regert *et al.* (1998), in these cases, extractions which hydrolyse polymerised or tightly bound compounds become necessary (Correa-Ascencio and Evershed 2014).

Although some of these limitations might be overcome by the use of adapted analytical techniques, a special effort must be made to detect and evaluate the products most prone to degradation. Recent research advancing the range of biomarkers available for plant residues (Colonese *et al.* 2017a, Heron *et al.* 2016 and Hammann and Cramp 2018) and case studies detecting their existence after careful analysis (Dunne *et al.* 2016, Spiteri 2012, Drieu 2017, Tarifa *et al.* 2019) indicate that organic residue analysis should not be regarded as an exclusively "meat reporting" technique for warmer and arid regions.

The process of domestication equally impacted animals and plants. Therefore, the possibility to shed new light in the social practices affecting both these products is of outmost importance to better understand a process in history which profoundly changed humankind. Nevertheless, one of the plant products which seem to present the best preservation in the environments studied in this PhD are resins, which, although they may played some role in prehistoric culinary practices acting as waterproofing agents and even possibly wine additives (McGovern *et al.* 1996, Garnier and Valamoti 2016), they may have also been used as glues. There is a significant body of evidence for the use of birch bark tar as adhesives (Rageot *et al.* 2015) in continental Europe accompanied by other types of resins which have been exceptionally detected (ex: Heron *et al.* 2015). Nevertheless, bitumen seemed to be the product of choice the Middle East for these same applications (Connan and Van de Velde 2010). This suggests that the knowledge of resin use might have not been widespread in the original Neolithic societies which left the Middle East at the onset of the neolithisation process. Whether they developed technology to produce birch bark tar independently or they took it after contacts with hunter gatherers using it might be difficult to prove. Yet, because hunter-gatherers did not use pottery, the presence of resin in this artefact proves the existence of a set of uses undetected in the Fertile Crescent and therefore unique to the first European farmers and pastoralists.

Birch bark tar was largely used either as a hafting adhesive or for repairing ceramic vessels in continental Europe during the Neolithic (Rageot *et al.* 2015), but pine resin seemed to be hardly used before the development of the Etruscan civilisation. Moreover, data supporting the use of conifer adhesives before the IVth millennia in the Mediterranean region seems to be scarce (Drieu 2017). Only five vessels from the greek Late Neolithic site of Mariyalos (Mitkidou *et al.* 2008) and one sample from Dikili Tash layers dating from ca. 4300 cal BC (Garnier and Valamoti 2016) report the presence of conifer diterpenoids. Evidences for the use of pine resins seem to be equally sparse beyond the meditterranean, with evidences in the French continental sites of Chalain and Clairvaux-les Lacs (Regert 2004, Masschelein-Kleiner 1989), one sample with dehydroabietic acid from Neustadt, Germany (Saul *et al.* 2013), one Neolithic globular amphora in Lithuania with a range of conifer diterpenoids (Heron *et al.* 2015), two samples with diterpenoid evidences in Skara Brae, Scotland (Muckerjee *et al.* 2008), and spectroscopic studies suggesting two samples in Runnymede Bridge (England) might have contained resin (Needham and Evans 1987).

The arrival of the Neolithic wave to the Iberian Peninsula opened new questions regarding adhesive use as birches were not available in such warmer climates. Although it would seem that, in the Mediterranean environment, resin from conifers could be the natural product more easily available to glue lithic tools into its hafts, there is an absence of compelling evidence proving this phenomenon due to the lack of surviving resins in the Cardial archaeological record. Only a hafting adhesive exceptionally preserved in the underwater sector from La Draga could be analysed under GC-MS. The results after a methylester derivatisation reported the presence of a monoterpenoid (borneol) and a series of diterpenoids including methyl abietic acid and degradation products such as methyl dehydroabietic acid and methyl didehydroabietic acid. Further phytolith and microcharcoal analyses suggested that the glue could have originated from a pinaceae species (Tresserras 2000b, Palomo *et al.* 2011). Nevertheless, other resin producing trees such as Juniperus sp. and Pistacia lentiscus were equally available in the forests of the Early Neolithic Iberian Peninsula (Revelles 2017, Riera *et al.* 2016).

Following the patterns suggested by Drieu (2017) and Rageot *et al.* (2015), evidences of pine resin in the Iberian Peninsula have been mainly reported from Bronze age argaric vessels. Two vessels from Peñalosa (Manzano *et al.* 2015), three vessels from La Bastida, one vessel from La Almoloya (Molina 2015b) and one vessel from Castro dos Ratinhos (García 2010) contained diterpenes interpreted as evidences of conifer resin by the authors. Nevertheless, the recent publication of two vessels containing the same molecular markers from layer IV in Cueva del Toro constitute the oldest evidence for the detection of pine resins in archaeological artefacts from the Iberian Peninsula and the whole Mediterranean (Tarifa *et al.* 2019). The paucity of the results after the publication of residues from more than 500 prehistoric ceramics, thus, suggest that the intensity of resin transformation in pottery vessels from the western

Mediterranean might significantly benefit from further evaluation. To date, all evidence is concentrated in the south of the Iberian Peninsula, a region with a distinct climatic situation, but this pattern could be significantly altered by performing new analyses from the remaining northern regions.

2.4 Questions regarding Early Neolithic culinary practices

Given that animal fats have been the most frequent type of lipid residue detected in prehistoric pottery, results could be used to explore meat culinary practices in the Neolithic. This perspective has usually been approached by comparing vessel contents and faunal records from the same site (ex: Copley *et al.* 2005d). Assuming no major biases selecting ruminants over non ruminants, or vice versa, the relative abundance of pig remains have been found to directly correlate with the intensity of non-ruminant isotopic signals in compound specific isotopic analyses of lipid residues (Mukherjee *et al.* 2007, 2008). This idea was put forward after the comparison of faunal and vessel content data from Durrington Walls, West Kennet, Wyke Down and Skara Brae in the British Neolithic. The researchers also noted that, despite a positive correlation with a 0.76 r^2 value, the relative quantity of non-ruminant fats in vessels was slightly under-represented when compared with relative quantities of porcine faunal remains. When taking into account Mukherjee et al's (2008) discussion, several factors might have been in place affecting this correlation:

1. One of the main assumptions was that bone assemblages and vessel residues would be comparable as they would originate from the same phenomenon, a set of cooking practices which would have resulted in these two types of remains. This meant that the lipids in the vessels must have been part of the animals whose bones had been found. Nevertheless, this assumption could be false if bones had been processed in a part of the site which was not accessed by the archaeologist. This might happen, for example, due to taphonomy problems or the lack of excavation of the right contexts as sites are not usually fully depleted unless necessary.

2. The evaluation of the culinary practices in pottery vessels might become significantly limited by small sampling sizes. Moreover, mixtures of different types of animal fats might render non-ruminant products invisible to the analytical methods available.

3. The existence of distinct culinary practices which would have treated ruminant and non-ruminant products differently. This might be related to the existence cooking techniques which may have not involved pottery (ex: spit-roasting), or rules or taboos rejecting the consumption of certain types of fats (Harris 1998).

The study of vessel residues from the British Neolithic performed by Mukherjee *et al.* (2007, 2008) produced an example of the interpretations of prehistoric culinary practices which might arise from organic residue analyses. They highlighted the low prevalence of vessels containing mainly pig products as ruminant signals seemed to dominate the assemblages. Nevertheless, further analyses of Grooved Ware pottery detected differences between the Durrington walls style and other northern pottery types and, most importantly, it detected a higher degree of processing and consumption of porcine products in no-habitat/ceremonial sites (Mukherjee *et al.* 2008), an idea which was revisited by Smyth *et al.* (2014) for causewayed enclosures in Ireland. Details on the culinary practices taking place in one of the aforementioned sites, Durrington Walls, have been further evaluated since thanks to additional analyses by Craig *et al.* (2015) and its findings have been a matter of interpretation and debate (Shillito 2019). In the opinion of this last researcher, the collection of porcine fats in vessels during spit-roasting (Albarella and Serjeantson 2002) constitutes an equally valid hypothesis which would explain the incidence of non-ruminant fats in pottery.

Regardless of the practices motivating the selective incorporation of one type of animal fats over the other, research on the British Neolithic provides an example of the discussions which may arise from focusing on the culinary practices in a particular period and region in prehistory. Following this line of research, more authors have explored the possible presence of cooking techniques where pottery is not involved. Copley *et al.* (2005d) suggested spit-roasting might have been practiced in pigs in the Eton Rowing Lake Neolithic site as these were 10% of the faunal record but were undetected in pottery residues. Šoberl *et al.* (2014) put forward the same hypothesis for Neolithic sites in the Adriatic sea and Cramp *et al.* (2019) reflected on the possible importance of spit-roasting in the Iron gates (Romania/Serbia) as the majority of pottery residues included biomarkers supporting the main transformation of aquatic foodstuffs.

Recent research therefore suggests that, although sites with direct correlations between porcine remains and non-ruminant lipid residues might be frequent, examples deviating from the correlation have been detected in both northern and southern Europe. One of these is Mala Triglavca, a rock shelter in Slovenia, where the disparity between porcine bones and pottery residues led the researchers to propose the existence of spit-roasting in the Mediterranean Early Neolithic (Šoberl *et al.* 2014). It is therefore of significant importance for this period to assess whether a disparity or a correlation can be built between porcine remains and non-ruminant lipids. Moreover, it would be significantly relevant for the understanding of both neolithisation routes to assess whether the social phenomena resulting in higher quantities of porcine fats in no-habitat/ritual sites in the final territories of the Danubian path is also present in the terminal region of the Mediterranean itinerary, the Iberian Peninsula.

Within this last region, caves are usually the sites most reporting no-habitat/ ritual activities. This includes ovicaprine pens (Bergadà 1998b, Bosch 2016b) and sepulchres in cavities such as Can Sadurní, Cova Bonica and Cova de l'Avellaner (Edo *et al.* 2019, Gibaja *et al.* 2018b), which concentrate the majority of the human remains so far recovered from the earliest Neolithic. Also, open air burials in the Bacelona plain include an adult woman from Plaça Vila de Madrid (Pou *et al.* 2010) and two infantiles from Carrer Reina Amàlia (González *et al.* 2011). Alternatively, open air sites have been found to present habitat-related features such as the hut basements in Guixeres de Vilobí (Oms 2014), Carrer Reina Amàlia (González *et al.* 2011) and Barranc d'en Fabra (Bosch *et al.* 1995a and b) and stilt architecture in La Draga (Bosch *et al.* 2000, 2011). Sites where the only structural evidences are underground silos might be more problematic to classify given that they do not offer direct evidences of settlement (ex: Caserna de Sant Pau, La Serreta, El Cavet) but would have certainly been in a location close to fields or other areas focused on the treatment of cereal products.

In essence, evaluating the possible presence of collective or private food consumption practices might offer insights into the social structure of the first farmers and shepherds of the Iberian Peninsula. The possible detection of deviations between animal residues and bone assemblages offers insights which might be complemented with other evidences. Detection of thermal alterations indicative of cooking at significantly high temperatures could be studied to explore the possible practice of frying or baking. Moreover, certain fungi and bacterial markers could be used to explore the possible intentional use of fermentation.

Beer and wine residues have long been one of the most elusive yet interesting types of residues studied by biomolecular archaeology. Far from being secure, identifications of this practice rely on inductive reasoning and contextual evidence which result in the interpretation that such practices might have been in place. Syringic and tartaric acid, biomarkers for grape, are usually invoked when assessing the presence of wine, yet archaeological analyses focused on this particular question have been mostly concentrated on ancient rather than prehistoric times (Michel *et al.* 1993, Pecci *et al.* 2017). Alternatively, beer and its origins remains a matter of intense debate amongst specialists. Calcium oxalates, a compound present in beerstone and a range of plants including spinachs (Spinacia oleracea), were detected in samples from the postcardial Neolithic and the bronze age in Can Sadurní (Blasco *et al.* 2008). Mead has also been the focus of interest of some researchers in the Iberian Peninsula, especially in the bell beaker period (Guerra 2006) yet no fully convincing analytical data has been able to push mead beyond the realm of speculative interpretation.

Regardless of the successes and shortcomings related to the detection of fermentation practices within archaeological samples, the origin of alcoholic beverages still remains one of the most public-engaging and interest-driving questions surrounding prehistory. The possibility that Neolithic societies ingested inebriating substances for ritual or recreational motives and the mere suggestion that their consequences might have even motivated humans to adopt agriculture (Hayden *et al.* 2012) is an indicator of the social importance these products have in the present and might have had in the past as a constant in human societies. Performing organic residue analyses capable of detecting fungal and microbial markers related with fermentation thus provides the possibility to obtain new data capable of understanding this integral part of present and past societies.

Nevertheless, such a cooking focused perspective on pottery residues could be seen as selectively ignoring the other half of the picture, uses not related with the management and consumption of food. The assumption that pottery had to be used for cooking has almost become a given of organic residue analyses and, therefore, something subjective of criticism. Is it possible that other practices were capable to result in the same molecular patterns nowadays attributed to food consumption? This question goes beyond the detection of non-comestibles such as resin or bitumen, and questions itself whether fats could be seen as something more than food. This is particularly relevant given that the processual tendency to model humans as stomachs with legs may prevent the exploration of a wider range of other possible pottery uses.

Treating fat as a combustible might be especially significant in contexts in need of artificial lighting such as caves or mines (ex: the middle Neolithic variscite mines in Gavà), nevertheless vessel shapes characteristic of lamps have been barely reported in the Early Neolithic. In consequence, in light of the wide range of possible uses pottery might have had at this period in prehistory, it is of outmost importance to approach its main sphere of use carefully. The use of vessels for cooking might be further supported by the existence of residues in specific parts of the vessel which have been related with boiling (rims) or thermal alterations in fats incompatible with alternative uses.

In conclusion, exploring prehistoric pottery use is a difficult endeavour limited by the necessity to assume that practices generating residues which could survive did effectively occur in the past. It is also affected by the possible existence of multiple social practices which might be completely undetectable or have unclear consequences. It remains a matter of zooarchaeological analyses to obtain direct evidences for spit-roasting, and the exploration of patterns arising from combining residues and animal bones is still in need of analyses on wider and more completely studied assemblages. Fermentation hitherto remains a highly controversial subject as waxes, resins, bitumen and other non-edibles are not the most commonly found and reported products. In essence, organic residue analysis are particularly well suited to help understand pottery use but they will not provide a clear solution to many archaeological questions related to pottery unless its results are properly integrated with additional lines of evidence. Analyses of samples from the north-east of the Iberian Peninsula at the onset of the Neolithic will be able to assess the presence of the biomarkers associated with these questions and, therefore, provide additional evidences of pottery containing edible matter which can be integrated into the interdisciplinary research system that is archaeology.

3 Methods

3.1 Biomolecular and isotopic analyses

As discussed by Evershed (2008), the analysis of archaeological organic residues in pottery relies on matching structures or distributions (chemical fingerprints), to compounds and mixtures known to exist in certain organisms. In this process, the potential of what could seem to be rather uninformative molecules can be greatly increased by taking into account the archaeological context. This section will therefore initially explore the principles and assumptions underlying the archaeological biomarker concept, the processes which may be involved in their incorporation into the clay matrix and the interpretative precautions which should be taken when interpreting an ancient residue. Once these more general notions have been established, the specific methods, analytical techniques and strategies applied in this research will be laid out.

3.2 Biomarkers in archaeology

3.2.1 Definition of archaeological biomarker and main classes.

Following Evershed (2008) archaeological lipid biomarkers could be defined as substances occurring in organic residues that provide information associated with a certain human activity in the past. The existence of families of fatty acids, alkanes, alcohols, esters, terpenoids and poliaromatic hydrocarbons can be linked to inputs from animal and plant fats, waxes, resins and bitumen. Moreover, the detection of hydrolysis, oxidation and reduction byproducts is of outmost importance to assess the degree of alteration of the studied residue and evaluate whether further studies will be able to provide useful archaeological information.

Moreover, isotopic ratios in pottery residues might help better understand the metabolic routes which generated specific compounds and thus achieve a better classification of the possible vessel contents. Lipids can be incorporated into animal tissues either via direct routing from dietary inputs or de novo synthesis after scrambling the ingested carbon into a common pool (Evershed *et al.* 2002b). Given that lipids are the most isotopicly depleted dietary components, de novo synthesis accessing the average isotopic dietary value will yield comparatively enriched fatty acids. In consequence, when different routes are used by the organism's biochemical processes to create fatty acids, changes in their isotopic values should be expected.

Given this situation, two main factors may explain the set of isotopic values acquired through organic residue analysis: environmental effects conditioning the $\delta^{13}C$ ratio before its incorporation into the animal organism (Mukherjee *et al.* 2005 and Keeling 1979), and phyisiological effects distributing, fractioning and synthesising fats once carbon has entered the digestive system (Everhsed *et al.* 2002b, Tieszen 1983, de Niro and Epstein 1977, 1978, Mukherjee *et al.* 2005, Cramp and Evershed 2014, Craig *et al.* 2012). For example, the distinct biosynthetic routes taking part in milk production have been shown to present a clearly distinctive pattern where stearic acid is at least -3‰ more depleted than palmitic acid (Figure 8).

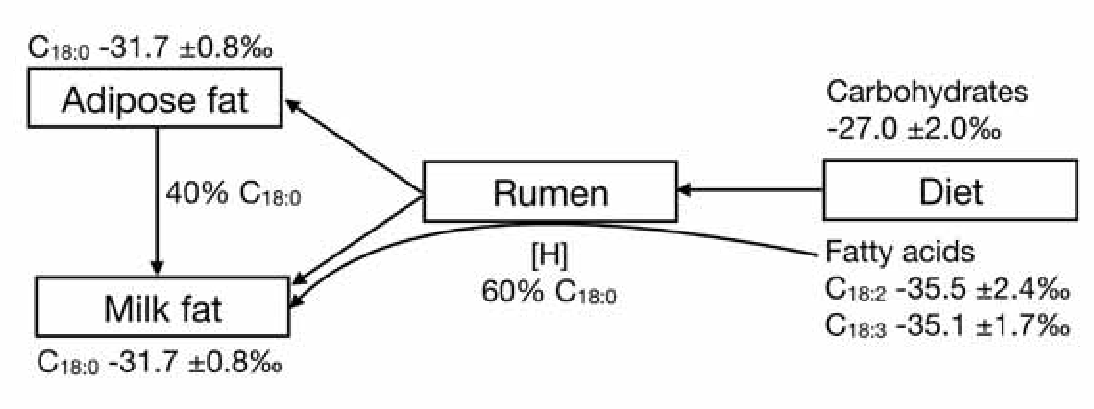


Figure 8: Diagram showing the routing of C18:0 fatty acids from the diet to adipose fat and the mammary gland through the rumen. Reproduced from Copley *et al.* 2003.

3.2.2 Incorporation of fatty acids into the clay matrix

It is assumed that the nature of the clay matrix would include pores both smaller and bigger than bacteria (Drieu *et al.* 2019). These cavities would contain lipids protected from microbial attack and they would de facto become the repositories of the organic matter analysed to understand vessel use. Once inside, residues would tend to distribute away from the inner surface of the vessel which was in direct contact with the contents (Stern *et al.* 2000). In consequence, samples from the inner surfaces have been considered to be more representative of use residues while extracts from outer surfaces would mainly contain contamination (Steele and Stern 2017).

Beyond vessel porosity, different substances seem to present a different transmission potential. As an example, only minimal quantities of grain lipids are incorporated while boiling but, when mixing with other fatty products, its incorporation improves (Hammann *et al.* 2018, Spiteri 2012). Additionally, milk could have provided a more appropriate substrate than only butter for the growth of bacteria, thus making the first product more prone to degradation (Copley *et al.* 2005a). Therefore, relatively high amounts of fat could also be the ones best suited for historic preservation.

Additionaly, the incorporation of lipids into the clay matrix seems to also be facilitated by the presence of a certain amount of heat. A rise in temperature would increase fat mobility, which, in turn, would help lipids permeate faster into the clay matrix (Charters *et al.* 1993b).

A common question present when interpreting organic residues is the determination of the range of uses that could have created the recovered lipids. Traditionally the explored solutions for this problem have been threefold. Firstly, it would be possible that the first use completely sealed the pottery and left no room for any further residue incorporation. Secondly, the reverse could be true and the last use could be the one present in the sample. A compromise between these two is the realisation that such a complex system would easily result in an average of uses trough the life of the vessel.

To explore whether this range of solutions makes sense, several effects observed over a range of experiments need to be taken into account. First of all, the rate of lipid incorporation might differ depending on the nature of the use. While the involvement of the vessel into culinary practices might spike the amount of lipids in the clay matrix, the presence of a cold product over a constant amount of time might suggest a slower incorporation velocity (Charters *et al.* 1993b). Furthermore, degradation experiments show that most of the incorporated lipids quickly disappear after a short period of time (Hammann *et al.* 2018, Hammann and Cramp 2018). Therefore, storage vessels and cooking vessels would undergo significantly different lipid incorporation and leaching dynamics. This seems to rule out that the vessel would contain only the first use of the vessel (unless the first was also the only use).

Alternatively, repeated use would slowly increase the amount of lipids into the clay matrix. Nevertheless, given that there is a limited amount of clay pores available, there is also a fixed limited amount of lipids which can be incorporated into the clay matrix. This maximum amount would also be different between more or less porous pottery fabrics. Once reached, only the degradation of part of the previous residue would allow the incorporation of organic matter from further uses. In the case of immediate reuse, barely any lipids could enter the vessel and, in cases of short reuse, only minor quantities of lipids would be incorporated into the clay matrix.

In conclusion, while the vessel is within its period of active use, it can be conceived as a complex system incorporating, degrading, and transporting the organic matter within the clay matrix. In this case, the most accurate option might be to treat archaeological residues as the averaged result of multiple uses since the beginning of vessel use.

3.2.3 Lipid degradation

After thousands of years buried underground, animal, plant or marine fats composed of any range of triacylglycerol species would become fully hydrolysed and broken down to their free fatty acid constituents. Polyunsaturated fatty acids will be seldomly detected (Evershed *et al.* 1992) given that a significant quantity of the unsaturated moieties would have been subjected to radical oxidation, yielding more water soluble compounds which would have been leached out. Finally, β-oxidation would have significantly contributed to the overall reduction of alkyl chain lengths. In consequence, the resultant would essentially be a range of free fatty acids with a minority of even chains whose ratios would not be identical to the ones found in the original triacylglycerides.

This does not provide an encouraging prospect for the study of the lipids recovered. Therefore, any interpretation of lipids preserved in pottery must be always informed by the possible degradation effects plausibly affecting the residue (Evershed *et al.* 1992).

Molecular biomarkers for degraded archaeological residues will rarely match those of modern references and, thus, a body of validation and degradation experiments and ethnographic studies gathered after more than 20 years of reseach (ex: Fox *et al.* 1995, Regert *et al.* 2001, Barnard *et al.* 2007, Evershed and

Charters 1995, Evershed *et al.* 1995b, Evershed *et al.* 2008a, Dudd 1999) has been necessary to understand the possible significance of the lipids recovered from pottery. This includes recalcitrant substances which may remain unaltered across time and molecules resulting from various degrees of oxidation and hydrolysis which still contain relevant information.

3.2.4 A case for lipids informing pottery use

Organic residue analysis assumes the presence of hydrophobic organic matter in pottery vessels. Given that lipids would be the molecules most easily preserved overtime, the recovered signal is not the one of a "complete" product or content, but just its partial remains. Therefore, it can also be assumed that, at the time the input took place, more molecule types accompanied lipids inside the pottery matrix. To interpret that vessels were specialized in the management of fat without further evidence would be akin to suggesting people was buried in skeletal form, which is a practice sometimes documented, but not widespread.

This fact is easily understood for degraded animal fats, which is usually assumed as evidence for the processing of the animal itself rather than the use of the fat for any specific purposes. The latter option is only a possibility that would need to be supported by further evidence. Alternatively, traces of animal fat could be explained by a multiplicity of activities, most of them related with cooking.

In conclusion, rather than taking lipids at face value, their presence should be also evaluated taking the range of pottery uses which makes archaeological sense in mind. Nevertheless, to reach this situation, samples must be carefuly selected, extracted and analysed. The following section unpacks the range of techniques which have been implemented in this PhD.

3.3 Applied methods and analytical techniques

3.3.1 Sample selection and preparation

Organic residue analysis aims at chemically characterising the remains of pottery contents that survived degradation. At present day, this is a destructive technique using mainly gas chromatography and mass spectrometry. Therefore, when selecting samples for organic residue analysis, some considerations have been taken into account:

- Location of the residue in the artefact: not all the parts of the pottery vessel contain the same quantity of residues. Had the object been used for boiling, the area close to the border would tend to contain higher quantities of lipids (Charters *et al.* 1993b). Alternatively, other types of culinary practices such as frying and stewing would promote the accumulation of higher quantities of fats in the base. Therefore, in cases where the base and the rim had been sampled, the type of use the vessel might have had will be better informed.

- The process of obtaining the sample: Given that the piece is perforated to obtain a certain amount of pottery, the analysis is completely destructive. Therefore, it is important to take the dimensions of the sherd into account as it needs to be able to withstand drilling. In this sense, pieces lighter than 10g tended to become problematic. In the case where, because of archaeological reasons, these were the only pieces which could be sampled, the only way to securely study them was by fully destroying the fragment. In this situation, the first millimetres from all the vessel's surfaces were not analysed but preserved in dust form.

- The selection of the pieces in its archaeological context: Samples were selected from diverse archaeological contexts to help perform comparisons and studies which would help better understand the artefact. Depending on the nature of the archaeological site, sample sherds were selected from several strata or from spaces functionally different. Moreover, different ceramic types were also selected when possible (open, closed and straight vessels for example), noting the presence or absence of decorations, the presence or absence of handles or other handling implements.

Samples were taken from 14 archaeological sites from the Cardial and Epicardial Early Neolithic periods in the north-east of the Iberian Peninsula. These included: El Cau de les Gulles, Berenguer de Palou, Carrer Reina Amàlia, Caserna de Sant Pau, Can Sadurní, Cova de Sant Llorenç, La Serreta, Guixeres de Vilobí, Cova de la Guineu, Cova de la Font Major, El Cavet, El Molló, El Barranc d'en Fabra and Cova del Vidre. When preparing samples, the most adequate techniques were based on a correct assessment of the nature of the studied vessel. Several options included:

A: Visible residues could be defined as an amorphous lump of assumed organic matter could be seen attached to the pottery surface. The colour of these residues would be mostly always black, although other colourations have been reported. Their texture is uneven and incorporates a network of micro-cracks. Only one relevant visible residue (Cova de Sant Llorenç) could be detected in pottery from the archaeological sites included in this tudy

B: Pottery matrix on big sherds: when no visible residues are present the study is carried out by partially drilling on the vessel's interior face. This is usually done by powdering a certain amount of pottery. This can be considered only partially destructive for the artefact given that the overall shape and morphometric characteristics will be preserved.

C: Pottery matrix on small sherds: when no visible residues are present and the sherds are significantly small and barely heavier than the minimal amount of pottery needed for analysis the whole sherd is fully destroyed.

Ideally, samples with the highest potential will be those selected on site and immediately stored at low temperatures. Although samples where adhered soil has not been removed yet are preferable, washed potsherds have also been shown to yield significant residues. This PhD has worked with pottery in a variety of conditions. While samples from some sites were recently excavated unwashed fragments (ex: Guixeres de Vilobí,), others had been cleaned and stored in museum collections and plastic bags for several years (ex: Barranc d'en Fabra). In all cases, the immediate interior surface was removed to remove possible post-excavation contaminants.

In order to further avoid modern contamination and cross contaminations between samples all its preparation was done in the cleanest environment possible. Nitrile gloves and safety glasses were worn at all times and the samples were preferably stored in aluminium foil in a cold environment. Photos were taken from the pottery sherd before and after extracting the sample so that the extent of the destructionwas documented. A basic database of the samples linking the vessel code with the sample code was of outmost importance as any confusion on the origin of the sample would lend the analysis completely useless.

3.3.2 Sample extraction

3.3.2.1 Sonication and silylation (solvent extraction)

The extractions were practiced on 2g of ground from the inner wall of the vase. The outside surfaces, which present higher probability of contamination (Heron *et al.* 1993), were removed before drilling. Ca. 5ml of Dichloromethane (DCM)/Methanol (MeOH) (2:1 v/v) were added to the sample. After a 15 minutes sonication and a 10 minutes centrifugation at 2000 revolutions per minute (rpm) the supernatant was extracted. This procedure was repeated 3 times and the three extracts were combined. The sample was then dried under a gentle stream of nitrogen, redissolved with DCM and derivatisatised with 100µl of N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% Trimethylchlorosilane (TMCS) at 40°C for 30 minutes. Finally, the sample was redissolved in DCM, transferred to a GC vial and spiked with 10µg of n-tetratriacontane as the internal standard (IS).

3.3.2.2 Acid methanolysis (acidified methanol extraction)

To asses possible losses during lipid preparation, samples from Cau de les Guilles, Caserna de Sant Pau, Berenguer de Palou, Can Sadurní, El Molló and Barranc d'en Fabra were spiked with 10µg of n-hexatratriacontane prior to extraction. Recovery values were found to be within acceptable ranges.

The extractions were performed on 1g of ground pottery. 4ml of methanol were added to the sample and the mixture was ultrasonicated for 15 min and acidified with 0.8ml of concentrated sulphuric acid. The result was heated at 70°C for 4 hours and then left to cool at room temperature. Lipids were extracted 3 times by adding 2ml hexane, vortexing, leaving the sample to partition, and extracting the hexane layer. The combined extracts were left on copper turnings over night and then dried under a gentle stream of nitrogen. Finally, samples were resuspended in hexane and transferred to GC vials, which had been spiked with 10µg of n-tetratriacontane as the internal standard. After screening for the presence of significant quantities of lipids, selected residues were trimehtylsilylated and submitted to chromatography for a second time.

3.3.3 Analytical parameters

3.3.3.1 GC-FID parameters

Institut de Ciència i Tecnologia Ambientals (ICTA-UAB):

1μl of each sample was injected in splitless mode to a 782A Agilent Gas Chromatograph fitted with a Flame Ionisation Detector (FID), and eluted trough an HP-1 capillary column (60 m length, 250 μm internal diameter, 0.25 μm film thickness) using hydrogen as the carrier gas. The oven temperature was initially set at 50ºC for 1 minute and then increased at 6ºC min^{-1} to 320ºC, where it stayed for 20 minutes.

3.3.3.2 GC-MS parameters

University of Bradford (UoB):

Samples were analysed with a 7890A Agilent Technologies Gas Chromatograph (GC) and a 5975C Agilent Technologies Mass Spectrometer with a triple axis detector and Electronic Ionisation (EI). Injection was done in splitless mode at a temperature of 300ºC and analyses were performed on an Agilent HP-5MS 30m x 0.25mm x 0.25 μm fused silica column with helium as the carrier gas. Oven temperature was set for an initial 2 minute isothermal period at 50ºC. Afterwards, it ascended at 10ºC min^{-1} until it reached a maximum of 350ºC. Temperature, then, was held constant for the remaining 10 minutes of the analysis. The resultant chromatograms were quantified and the eluted compounds were identified using its mass spectra and compared with a reference database (NIST 2.0).

The British Museum (BM):

The analyses were performed with a 6890 Agilent GC equipped with a HT5 column (12m x 220 μm x 0.1 μm). The sample was injected on-column with the oven temperature at 50ºC. After a two minute isothermal period, themperature rose at a rate of 10º/min until it reached 320ºC. This one was kept constant for the final 15 minutes of the analysis. The GC was coupled with a 5975c Agilent Mass Spectrometer equipped with an electric ionisation source and a simple quadrupole operating in full scan mode between *m/z* 50 and *m/z* 750. The detected molecules were identified using the NIST2.0 library.

ICTA:

From the extract in isooctane solution, 1µl was injected into an Agilent 7890A GC coupled to an Agilent 5975C Mass Spectrometer. The GC was fitted with a DB-5MS column measuring 30m x 250µm x 0.25µm. The GC injector was operated in splitless mode and helium was used as the carrier gas. The oven temperature was set at 50ºC for 2 minutes and then increased at 15ºC min^{-1} until it reached 170ºC, then a new ramp at 5ºC min^{-1} brought the oven to 320ºC and held at that temperature for 6 minutes. The Mass Spectrometer was run in electron impact mode and masses were acquired in full scan mode between m/z 50 to m/z 800.

3.3.3.3. GC-C-IRMS parameters

In order to correct for the methylation of the carboxyl group, a C16:0 and C18:0 external standard was prepared with 100 mg of the compound in 100ml of hexane (UoB and BM) or isooctane (ICTA-UAB) . An aliquot of 100 µl was extracted using the acidified methanol procedure and analysed by GC-MS to obtain the retention times of the aforementioned molecules. External standards were prepared for every batch of samples.

Liverpool University:

Carbon stable isotope ratios were determined using a Delta V Advantage isotope ratio mass spectrometer (Thermo Fisher Scientific, Bremen, Germany) linked to a Trace Ultra gas chromatograph (Thermo Fisher) with a ConFlo IV interface (Cu/Ni combustion reactor held at 1000ºC; Thermo Fisher). All samples were diluted in hexane and 1µl of solution was injected into a DB5 fused-silica column (30m x 0.25mm id x 0.25µm film thickness). Following a 1min isothermal period at 45ºC, the oven temperature was raised by 6ºC min^{-1} to 295ºC and held there for 15 min. Helium was used as a carrier gas and the eluted products were combusted to CO_2 and ionised in the source of the mass spectrometer by electron ionisation. m/z 44, 45 and 46 were monitored to compute the $^{13}C/^{12}C$ ratio of each peak. Pulses of reference CO_2 gas were injected and were used for comparison using the Isodat 3.0 Gas Isotope Ratio MS Software (version 3.0; Thermo Fisher). Instrument precision was <0.3‰ and accuracy was <0.5‰.

ICTA:

Compound-specific isotopic analyses were performed on a Delta V Thermo Fisher isotope ratio mass spectrometer linked to a Trace GC Thermo Fischer Scientific. Helium was used as the carrier gas and the combustion reactor was set at 940ºC. Samples were diluted in isooctane and between 0.5 to 2 µl of solution were injected into a DB-5 MS (60m x 0.25mm x 0.25 µm) column depending on the sample's fatty acid concentration. The injector temperature was set at 310ºC and the oven was initially set at 80ºC for 1 minute, then ramped at 30ºCmin^{-1} to 120ºC, then increased the temperature to 320ºC at 6ºCmin^{-1} and held in this setting for and additional 21 minutes.

In both cases results have been presented in the standard notation relative to the Vienna Pee Dee Belemnite (V-PDB) standard. $\delta^{13}C$‰ = [(Rsample-Rstandard)/Rstandard].

Values were further corrected to account for the methylation of the carboxyl group. Mass balance correction and simple correction were used (Rieley 1994). Both methods present minor variations in its results, nevertheless, values from the mass balance correction have been considered more robust and have been used for residue interpretation.

3.3.3.4 EA-IRMS parameters

University of Bradford:

5mg of coating from samples 8726 and 7778 and two calcium carbonate crusts from fragment 7778 were extracted and ground in a zionite mortar. 1mg from each was accurately weight and placed in a tin capsule. Samples were analysed by IRMS using an elemental analyser linked to a PDZ Europa 20/20 mass spectrometer. This technique has detected the percentage abundance of Carbon and Nitrogen in the sample and has calculated its delta isotopic values. A two point linear normalisation was performed with laboratory internal standards (Paul *et al.* 2007).

The selection of analytical strategies applied in this PhD was also informed by two small case studies aimed at better understanding the extraction processes to be performed.

3.4 Studies informing the analytical strategy:

3.4.1 A comparison between the acidified methanol extraction and the solvent extraction

To test the degree of lipid preservation, 10 samples and two possible visible residues from layer 59 in the Carrer Reina Amàlia (Barcelona) archaeological site (see section 4.3 for further details) were selected to test preservation rates by comparing the results of the solvent extraction and the acidified methanol extraction.

Sample	Total Lipid Extract ($g{\cdot}g^{-1}$)		Percentage increase	Shape
	DCM/MeOH	Acidified MeOH		
8392	0	20	-	Base
6522	4	21	416%	Rim
8842	0	35	-	Body
7778	0	115	-	Rim
6231	161	319	98%	Rim
8726	162	214	32%	Rim
5132	162	432	167%	Rim
6681	201	374	86%	Rim
8737	216	1015	370%	Rim
5531	306	428	40%	Rim

Table 4: Total lipid extracts from solvent and acidified methanol extractions and the percentage increase in their yields.

The quantification results (Table 4) show that the acid extraction consistently results in higher lipid yields. The DCM/MeOH extraction yields more than $5\mu g{\cdot}g^{-1}$ in 60% of the samples, whereas in the acidified methanol extraction 100% of the potsherds exceed this threshold.

Figure 10 show the case of sample 8737. As depicted, C16:0 and C18:0 fatty acids seem to be the most abundant molecules. Nevertheless, other significant compounds such as saturated (Cm:0), unsaturated (Cm:1) and branched (Cm:0br) fatty acids have been detected. For a detailed report of all the detected molecules, elution times and abundances, see the appendix 3.

In terms of the detection of molecules, Table 5 presents the compounds that, in this case, have been found in only one of the two extraction types.

DCM/MeOH extraction	Acidified Methanol extraction
Glycerol	Very long chain fatty acids
Short chain fatty acids	Dicarboxylic acids
Monoacylglycerols and Diacylglycerols	Ketoacids
Squalene and Cholesterol	C14:1, C22:1
Underivatised molecules	C19:0br
Short chain alcohols	ω-(o-alkylphenyl)alkanoic acids

Table 5: Chemical compounds detected only in one of the two extraction types.

Molecules attributed to contamination have been detected in both methods. Squalene may have their origin in human skin lipids and could have been embedded in the samples due to handling by the archaeologists. Phthalic acids are common constituents in plastic bags and are highly volatile. Therefore, long term storage in such containers could be the origin of these molecules. Since the source is clearly not ancient, the amount of molecules attributed to contamination has not been measured.

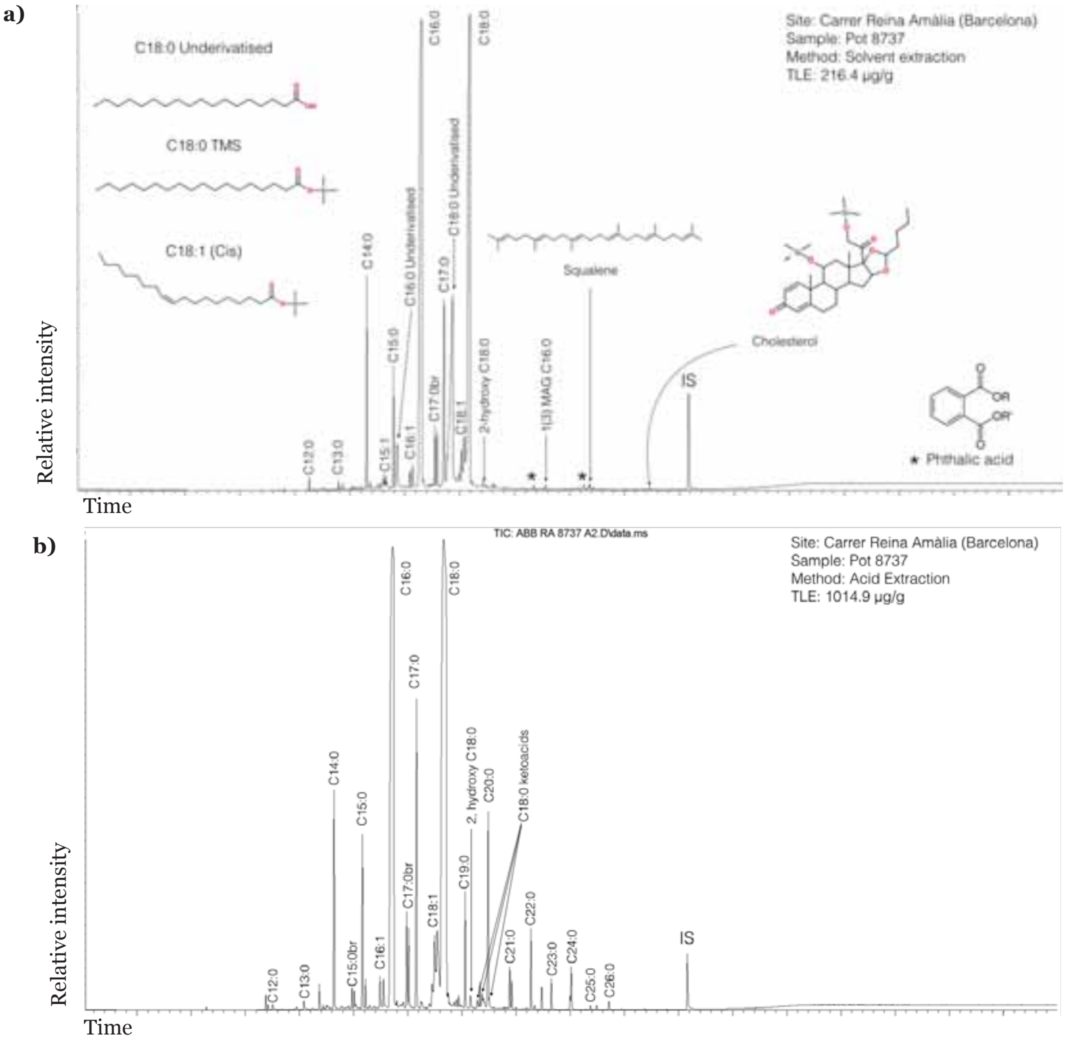


Figure 10: Representative chromatograms of total lipid extracts from solvent and acidified methanol extractions

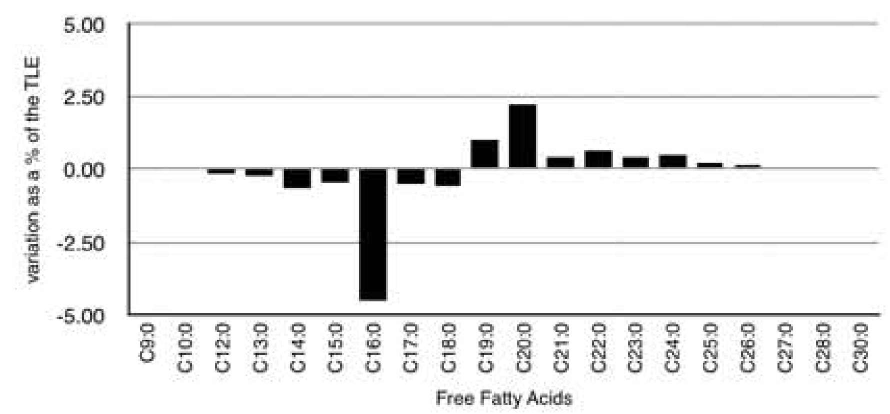


Figure 9: Aveaged variation in the lipid yield of saturated fatty acids from the 10 compared samples. Negative values reflect the prevalence of a given fatty acind in the solvent and positive values reflect higher yiels in the acidified methanol extraction.

In terms of relative quantities, almost all compounds that are found in both methods present minor variations. In percentage of the TLE, only three molecules reveal significant differences. The major change occurs in the C16:0 fatty acid. Its relative abundance decreases by 4.5% in the acidified methanol extraction. Alternatively, the C18:1 and C20:0 fatty acids present a 2% increase. Figure 9 presents the variation of the saturated fatty acid profile. Underivatised and derivatised fatty acids with the same chain lengths have been considered the same molecule.

As presented, shorter chain fatty acids seem to be slightly less abundant in the acidified methanol extraction. This phenomenon is completely reversed when chain lengths are longer than 18 carbons.

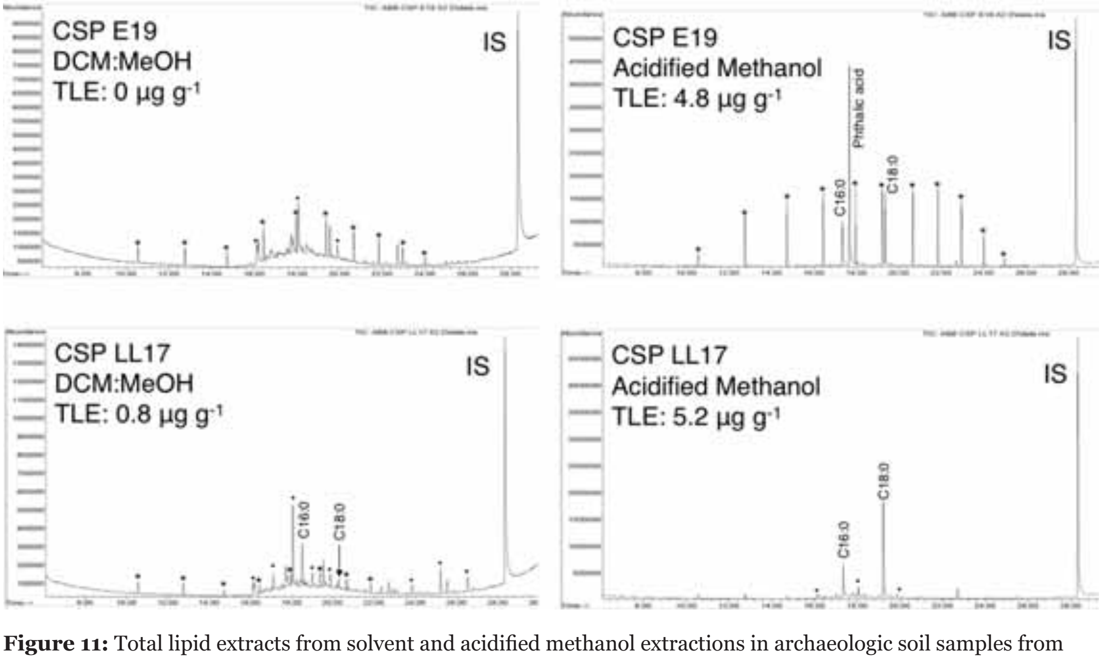


Figure 11: Total lipid extracts from solvent and acidified methanol extractions in archaeologic soil samples from Caserna de Sant Pau (Barcelona).

Finally, two soil samples from the Caserna de Sant Pau site were analysed with both DCM/MeOH and acidified methanol extractions. The goal of this analysis was to check for possible soil contamination from the region. Although not coming from the same archaeological site, Caserna de Sant Pau is situated just across the street from Carrer Reina Amàlia and therefore it can be argued that samples share the same geological and environmental characteristics. Chromatograms for both extractions are presented in Figure 11 and present no significant amounts of lipids. Therefore, contamination from soils should be considered minor if this data is taken into account.

The results of the extraction study show that both the acidified methanol and the DCM/MeOH extractions have been able to recover significant amounts of lipid from the vessels found at Carrer Reina Amàlia. In the case of the solvent extracts, triacylglycerols are clearly absent. Trace amounts of diacylglycerols have only been detected in one sample, some monoacylglycerols seem to be widely present in trace quantities and, in some cases, free glycerol has been found. With the detection of oxidation products, this profile clearly shows the presence of extensive oxidation and hydrolysis that has broken down the original fatty compounds. In consequence, the lipid profiles are only composed of free fatty acids of different chain lengths. This is not the optimal situation in terms of the identification of the residue, but it means that significant amounts of lipids can be recovered in western Mediterranean environments. Furthermore, results from a base and a body sherd yielded low amounts of lipids. As it has been previously reported by Charters *et al.* (1993b), this suggests that the rim is the area in which cooking pots absorb the most lipids.

When studying which method might be more effective in this case study, it can be argued that an increase of lipid yields in the acidified methanol extraction, far from compromising the results, has helped to improve them due to three main reasons.

(1) The acidified methanol extraction has recovered significant quantities of lipids where the DCM/MeOH extraction has failed. Therefore, the acidified methanol extraction provides a chance to improve the result from organic residue analyses in more dry and hot parts of the world, such as the Barcelona environment.

(2) An increase in the amount of lipids recovered offers the opportunity to reduce the amount of pottery needed for the analysis. In this study, the DCM/MeOH procedure has been executed on 2g of ceramic, whereas the acidified methanol extraction has been performed on just 1g and yet a mean 145% more $\mu g \cdot g^{-1}$ of lipids were recovered.

(3) The acidified methanol extraction generates methyl ester derivatives as part of the extraction process. There might have degraded any previously complete triacylglycerols. Given that these did not survive in the first place, it can be argued that most of the interpretative potential offered by the solvent extraction was already lost due to the aggressive degradation processes occurring in Mediterranean environments. Moreover, other partial losses of information due to absence of alcohols can be solved by silylating the OH groups before analysis, which would provide a means of detecting the possible hydrolysis of wax esters, as suggested by Correa-Ascencio and Evershed (2014).

3.4.2 Visible residues from calcium carbonate crusts

In some cases, the existence of some crusts possibly originating from pottery use was detected in samples from CRA. Calcium carbonate crusts are commonly found in the northeast of the Iberian Peninsula given the abundance of soils incorporating this type of rock. To understand their potential, two dark pottery linings found in the interior surface of the vessel and two samples of calcium carbonate were studied using Isotope Ratio Mass Spectrometry (IRMS).

Id	Sample	Surface	Material	N (%)	C (%)	$\delta^{13}C$‰
1	7778	Exterior	Calcium carbonate	0.0	6.1	-11.3
2	7778	Interior	Calcium carbonate	0.0	7.5	-11.3
3	7778	Interior	Possible residue	0.0	1.5	-15.4
4	8726	Interior	Possible residue	0.1	2.7	-26.5

Table 6: Results from elemental and $\delta^{13}C$ isotopic analyses of crusts adhered to the interior surface of pottery from Carrer Reina Amàlia.

As it can be seen in Table 6, the nitrogen values in the four samples are lower than 0.1%. This fact suggests an absence of amines, nitrates and other organic molecules with nitrogen in its chemical composition. Nonetheless, the carbon values vary between 1.5% and 7.5%, which suggest a small presence of chemical compounds that would have this element in its chemical structure.

Carbon is a chemical element that, in some cases, can be found in inorganic matter (carbides, carbonates, cyanides and carbon oxides). In order to assess whether the quantity of detected carbon could have been part of an organic residue, the results have been compared with data in the literature. More specifically, three extensive studies on visible residues from northern Europe have been used. These papers have followed the same technique applied in this research (Campbell *et al.* 2004, Craig 2004, Spangenberg *et al.* 2006). Nevertheless, the cold and wet environment in Belgium, Switzerland and Scotland makes preservation of organic matter easier. Therefore, organic residues from the Mediterranean could present different values due to poorer preservation of organic matter. Unfortunately, the absence of specific literature on the application of this technique in southern Europe makes comparison only possible with data from the north.

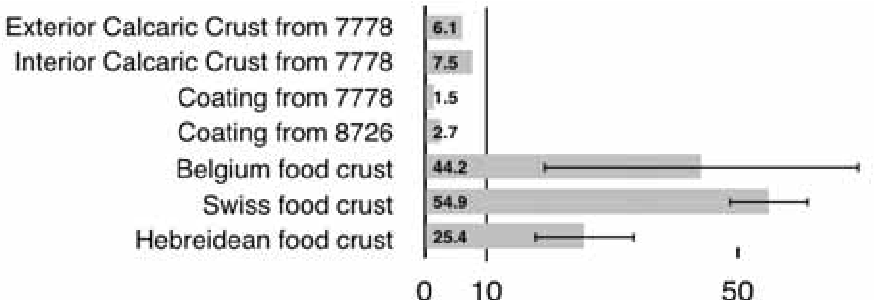


Figure 12: Quantity of carbon in the surface crusts is compared with reference values from archaeological food-crusts in northern Europe. Sources: Spangenberg *et al.* 2006, Craig 2004, Campbell *et al.* 2004.

As depicted in Figure 12, the carbon quantities of the two possible residues are much lower than the values which visible food crusts normally present. Nevertheless, these values could be low because of poor preservation and the presence of a certain quantity of inorganic material from the ceramic matrix. Furthermore, it is important to notice that both calcium carbonate crusts present higher quantities of carbon than the black coatings. Therefore, the likelihood of being of mineral origin should not be ruled out. To further assess these concerns, the isotopic values have been taken into account.

As shown in Figure 13, although one of the samples falls within the rage of the expected values in the literature, it would be risky to treat it as a visible residue because of the low percentage of carbon. Nevertheless, its isotopic values are similar to what can be expected from this type of material (Oehlert and Swart 2014), which suggests that did it contain relevant information related to pottery use, further significant research will be necessary until their potential is fully understood (Hendy *et al.* 2018).

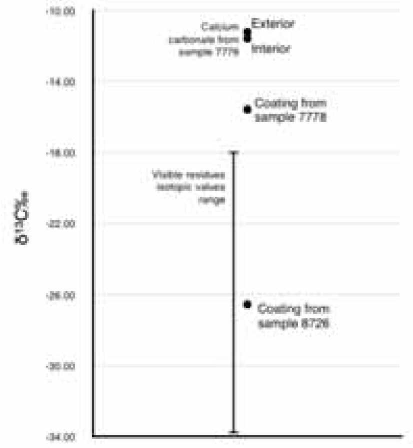


Figure 13: $\delta^{13}C$ isotopic values obtained from the analysed crusts. Reference values for visible archaeological residues were obtained from: Craig *et al.* 2002, Heron *et al.* 2015, Campbell *et al.* 2004, Craig 2004 and Spangenberg *et al.* 2006

Stable isotopic studies on the C16:0 and C18:0 fatty acids were based on a significantly different approach than the one practiced for the study of said visible residues. In this case, Bayesian models proved to be an important tool in systematically assessing the possible origins of the recovered animal fats.

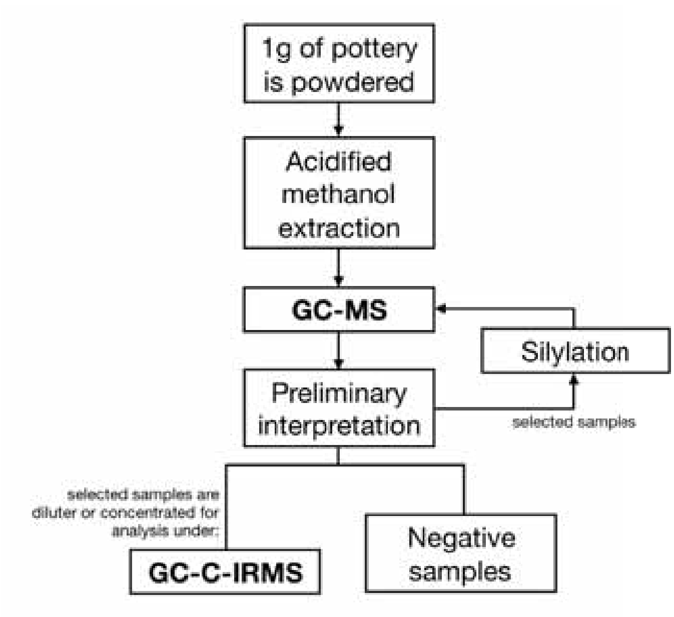
3.4.3 Analytical strategy

In order to obtain molecular and isotopic data from a significant quantity of archaeological sites across the northeast of the Iberian Peninsula, samples were analysed in different laboratories. Molecular analyses from Carrer Reina Amàlia were performed at the University of Bradford and compound-specific isotopic analyses were completed at Liverpool University and the Institut de Ciència i Tecnologia Ambientals (ICTA) in the Autonomous University of Barcelona. Other archaeological sites including Cova de Sant Llorenç, Caserna de Sant Pau, Can Sadurní and Cova del Vidre were studied in the British Museum laboratories and later compound-specific isotopic values were performed in the ICTA facilities. The remaining sites (Cau de les Guilles, Berenguer de Palou, La Serreta, Guixeres de Vilobí, Cova de la Guineu, El Cavet, Cova de la Font Major, El Molló and Barranc d'en Fabra) were completely analysed at the ICTA laboratories.

Analytical strategies adapted to the instrumentation available in each facility were put in place. Figure 14a details the procedures applied in the University of Bradford and the British Museum while Figure 14b presents the sequence followed in the ICTA laboratories.

In all cases, compound-specific isotopic data was later treated using mathematical mixing models in order to assess the possible ranges of animal products contained by the vessels.

a)



b)

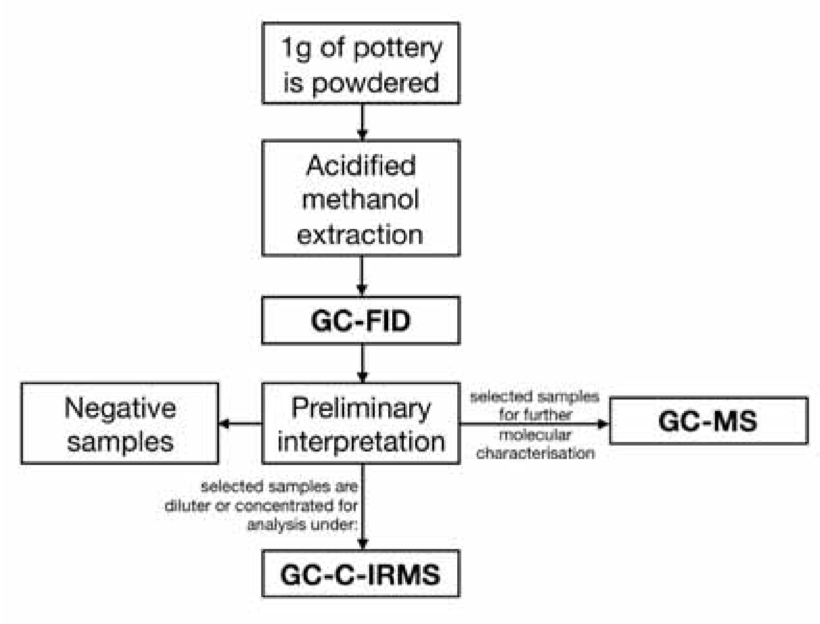


Figure 14: a) sequence followed in the University of Bradford and the British Museum. b) Sequence followed at the ICTA-UAB facilities

3.5 Mixing models in organic residue analysis.

Pottery found in domestic contexts would have probably had more than one use. Because the diet is composed of a range of foodstuffs, it is difficult to expect that vessels would have been consistently used to process only one type of product. Moreover, any recipe containing a number of different fat-bearing ingredients would generate a residue resulting from its mixture. Therefore, when interpreting the nature of organic residues embedded in the clay matrix, the presence of a mixture of products should always be assumed unless there is specific data available which indicates otherwise.

The evaluation of the presence of certain biomarkers helps identify the presence of a certain product but does not exclude the possible existence of other foodstuffs. Nevertheless quantitative analysis of fatty acid isotopic ratios must evaluate the possible impact the mixing of multiple products might have had. To date, several mixing models working with modern reference data have been used to provide an approximate value of the weight of each possible product in the resulting fatty acid isotopic values.

The first contribution to this problem (Dudd and Evershed 1998) calculated mixing curves previously used for the study of vegetable oil adulteration (Woodbury *et al.* 1995). The following formula was applied to obtain theoretical isotopic values from mixing given percentages of two types of fats (A and B) (Mukherjee *et al.* 2005).

$$\delta^{13}C_{mix} = \delta^{13}C_{(A)}\left[\frac{(X \cdot A)}{(X \cdot A) + (Y \cdot B)}\right] + \delta^{13}C_{(B)}\left[\frac{(Y \cdot B)}{(X \cdot A) + (Y \cdot B)}\right] \quad \mathbf{1}$$

This equation (x) includes: δ^{13}Cmix, the predicted isotopic ratio for a fatty acid after mixing two different fat sources (A and B); δ^{13}C(A), the δ^{13}C values from the centre of the distribution of modern reference values in fat A; δ^{13}C(B), the δ^{13}C values from the centre of the distribution of modern reference values in fat B; X, the amount of type A fat present (in %); Y, the amount of type B fat present (in %); A, the percentage of the specific fatty acid in type A's triacylglycerols; and, B, the percentage of the specific fatty acid in type B's triacylglycerols.

The successful application of this approach to diverse archaeological studies (see Figure 15) (ex: Dudd 1999, Evershed *et al.* 2002a and 2002b, Mukherjee *et al.* 2007, 2008, Molina 2015b, Colonese *et al.* 2017b), revealed the necessity to work with more sophisticated models to overcome several limitations:

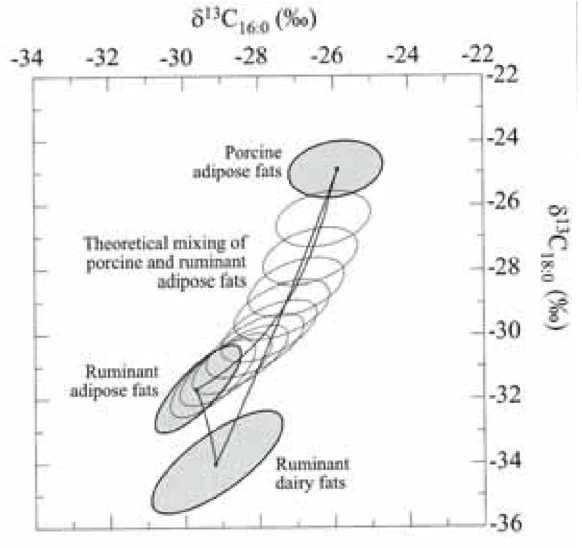


Figure 15: Application of theoretical mixing curves to reference dairy, ruminant and non-ruminant adipose fats. Reproduced from Muckherjee *et al.* 2005

- Mixing curves could only study possible mixtures between two products. This was therefore mainly used to study percentages of ruminant and non-ruminant adipose fats without the ability to evaluate possible inputs from third or fourth sources such as milk, vegetable or marine lipids. Moreover, the addition of n isotopic signatures would able to resolve the contribution of n+1 sources but, should the number of possible products in the mixture be higher, the model would become mathematically undetermined (Phillips and Gregg 2003).

- Mixing curves did not allow for any uncertainty measures such as margins of error in the measurement of isotopic values or standard deviations resulting from the study of the distribution of modern reference samples. Nevertheless, substantial variability between lipid sources, variation within modern reference values and small analytical measurement errors should be taken into account given its possible influence in the final interpretation (Phillips 2012). Failure to do so would, for example, generate artefacts where values plotting within the expected values for a given lipid source would still be considered the result of a mixture.

- Mixing curves only provided a single line in which the values measured in archaeological pottery would be coherent with the expected $\delta^{13}C$ ratios for a given mixture. Therefore, the evaluation of any data point outside the curve was still dependent on the subjective interpretation of xy plots.

After significant work by Phillips (2012), Phillips and Gregg (2001, 2003), Phillips *et al.* (2005, 2014), Parnell (2010, 2013), Fernandes (2014, 2015) and many others, new, more complex and refined mathematical approaches are now capable to study mixtures of more than n+1 sources where both the reference data and the results are treated as probability distributions. Results include the probability a certain source contributed a given percentage in the mixture and they can be studied statistically to reject or maintain null and alternative hypotheses.

After the work of Fernandes *et al.* (2017), a specific implementation of said mixing models has been adapted to the specific necessities of the study of fatty acid isotopic ratios from organic residues in pottery. "Food Reconstruction Using Isotopic Transferred Signals" (FRUITS) is a software for Bayesian modelling aimed at diet reconstruction (Fernandes *et al.* 2014) capable of working with informative and uninformative priors and equipped with parameters which take weighted contributions into account. In essence, it uses the same mathematical principles as other Bayesian models (Dirichlet distributions, Gibbs sampling and a Monte Carlo Markov Chain) such as MixSIAR (Stock *et al.* 2018) in a graphic user interface independent of command based software such as R, which provides a smoother learning curve. FRUITS, then, can generate a BUGS (Bayesian inference Using Gibbs Sampling) script which runs using the OpenBUGS software and returns both raw probability distributions in text and a range of graphs to better interpret the results. Studies concerning organic residue analyses have successfully applied FRUITS' models into case studies from Japan (Lucquin *et al.* 2018), England and Germany (Fernandes *et al.* 2017). These have been able to provide evaluations on the degree of ambiguity existent in interpretations based on fatty acid isotopic ratios and have revealed patterns in pottery use which would have otherwise remained unknown. In conclusion, these Bayesian mixing models offer the opportunity to perform more nuanced interpretations of vessel use provided that the link between the archaeological questions and the characteristics of the model is well established.

In conclusion, given its user friendliness and capability to work with the most advanced mixing models to date, version 3 of the FRUITS software (Fernandes *et al.* 2017) has been used in this PhD to obtain estimates for the contribution of different sources into the measured isotopic values from palmitic and stearic acids in organic residues from pottery use.

3.5.1 Bayesian mixing models.

It is not the goal of this PhD to demonstrate a comprehensive understanding of the mathematical processes underlying Bayesian inference, but it is always necessary to understand the nature of the techniques applied to the studied data as much as possible in order not to fall into "black box" situations. Therefore, this section will initially describe the main mixing models used in organic residue analyses and their core characteristics. After the necessary notions have been laid out, the second part of this section will present the mathematical models used in this PhD. For more detail, further reading can be found in Phillips and Gregg (2001), Parnell *et al.* (2010, 2013), Fernandes *et al.* (2014, 2017), Stock *et al.* (2018) and Lucquin *et al.* (2018, see appendix within).

As described by Fernandes *et al.* (2014, 2017), the model in FRUITS follows, in its core, the same principles applied by Woodbury *et al.* (1995): the value resulting from the mixture of a range of sources is a weighted mean including the values of said sources, the contribution of each source and the concentration of the fatty acid in the source's triacylglicerides. Nevertheless, going beyond Woodbury's implementation, FRUITS includes new parameters to account for external factors affecting the mixture (such as the Suess effect) and treats some parameters statistically to account for measurement errors and the natural variability within fat types.

$$\mu_{H,k} = \frac{\sum_{i=1}^{N_k} \alpha_i C_{ik} (T_{ik} + I_{ik})}{\sum_{i=1}^{N_k} \alpha_i C_{ik}} \qquad 2$$

This is the formula (2) reproduced from Fernandes *et al.* (2017), which includes: N, the number of lipid types; μH_k the mean of each measured isotopic signal; α_i, the unknown contribution of each type of fat (i) which will be calculated; C_{ik}, the concentration of a given fatty acid (k) in the triacylglycerides of a certain type of fat (i); T_{ik}, the offset due to fluctuations in atmospheric $\delta^{13}C$ values in each fatty acid (k) in a certain type of fat (i); and, I_{ik}, the modern reference values of each fatty acid (k) in each type of fat (i).

In essence, for every fatty acid (palmitic and stearic acids) measured in each fat type (ruminant adipose, ruminant dairy, non-ruminant adipose), the summation of the unknown contribution times the concentration of said fatty acids in the tryacilglycerides times the addition of the modern reference values and its offset is divided by the summation of the unknown contribution times the concentration of said fatty acids in the triacylglycerides. This equals the mean of each fatty acid for each type of fat.

$$P(H|E) = \frac{P(E|H) \cdot P(H)}{P(E)} \tag*{3}$$

Nevertheless, this formula alone does not provide a mechanism for treating the results of the mixing model as probability distributions. To that end, Bayes's theorem must be applied (Bayes 1763). In a nutshell, it states that given the probability of a hypothesis (H) being true, this can be updated after considering the probabilities associated with new detected evidence (E), which is expressed in the following formula (3):

Following the expression (3), the probability of a hypothesis given that certain related evidence has been detected P(H|E) equals the probability of finding such evidence given the hypothesis P(E|H) times the probability of the hypotheses itself P(H) divided by the probability of finding said evidence regardless P(E).

When using bayes's theorem, the first term in the equation (P(H|E)) is considered the posterior probability, the first term in the numerator (P(E|H)) is named as the likelihood and the second term (P(H)) is the prior probability. Therefore, it can be stated that the combination of prior beliefs with a certain dataset can result in posterior believes. The Bayesian approach has been already used in archaeology to solve a range of problems but maybe the most common one is its application to ^{14}C dates (Buck *et al.* 1996). In that case, stratigraphy can be used to establish a prior probability which, when combined with the measured ^{14}C values, provides a posterior probability with a more precise absolute dates (Ramsey 2009)(Ortman *et al.* 2007).

In the case of the Bayesian mixing model applied in this PhD, the characteristics of the priors, the likelihood and the techniques used to obtain the posterior distributions are described below.

3.5.1.1 Priors

Bayesian models usually incorporate two types of priors. Informative priors bear information provided by the researcher which will affect the interpretation of the posterior and uninformative priors are the descriptors of the statistical distributions whose values do not affect the interpretation. It is therefore possible to create a Bayesian model without any significant prior information, but uninformative priors must always be well understood in order to take its limitations into account when interpreting the posterior probability.

In the case of this mixing model, the unknown parameter (the contribution of each source) is modelled as an uninformative prior in the shape of a Dirichlet distribution (Fernandes *et al.* 2014). The Dirichlet function assumes each source is present in the mixture and, therefore, the model searches for a quantity higher than 0 that fits the provided data. In consequence, the more sources the model is given, the less abundant each source is assumed to be individually. This effect is due to the fact that, in order not to be informative, each source has to be considered equiprobable. Therefore, if the model contains three sources, it is assumed that each will have an expected abundance of 1/3, but when 10 sources are incorporated, the prior will assume an abundance of 1/10 for each. This limitation implies that the number of included sources has to be carefully evaluated as it will have some effects in the results. In essence, before a specific product is included in the model, there should be archaeological evidence supporting its presence in the diet and within the vessel residue. For example, the inclusion of strawberries as a possible source in the mixture will bring the model to report that, at least, a minimum amount of strawberries were part of it. If that is the only archaeological evidence supporting the presence of strawberries, the argument becomes circular and, moreover, the predicted amount of other products will have been underestimated.

When data suggesting some sources were more probable than others is available, the prior parameters can be modified to reflect this fact. In the case of organic residues, the presence of certain biomarkers, bioarchaeological data, pollen records and other studies which may help infer the range of available products in the site and its importance could be used as informative priors. In the research performed by Lucquin *et al.* (2018), priors giving more weight to aquatic products were put in place when certain biomarkers were detected (presence of phytanic acid, abundance of the SSR phytanic acid diastereoisomer and the presence of C20 APAAs).

3.5.1.2 Likelihood

According to Fernandes *et al.* (2017), formula (2) is used as the likelihood function with all the data provided by the user (sources and value of the organic residue). Compared with other uses of FRUITS in diet reconstruction from C and N isotopes in collagen, when assessing mixtures of fatty acids, the characteristics of several parameters might change.

Firstly, given that fatty acid concentrations in the triacylglycerols and δ^{13}C16:0 and δ^{13}C18:0 are highly correlated values which present a certain degree of inter-individual heterogeneity and possible measurement errors, it is better to conceive them as multivariate normal distributions with a mean vector and a variance-covariance matrix (Fernandes *et al.* 2017).

Moreover, in the case of lipid residues, many possible solutions could account for the observed data because of the proximity of different sources and the fact that these spread along certain lines (co-linearity). To paliate these limitations, Fernandes *et al.* (2017) proposed to use the Standard Error of the Mean ($\mathrm{STE}_{\mathrm{mean}}$) as the value representing the error in the distribution of the source data. The standard error of the mean can be conceived as an estimate of how far the sample mean could be from the original mean and it is calculated by dividing the standard deviation (σ) by the square root of the number of observations (n) (4).

$$\mathrm{STE}_{\mathrm{mean}} = \frac{\sigma}{\sqrt{n}} \tag{4}$$

3.5.1.3 Posterior

The obtention of the posterior distribution is performed using several algorithms (The Markov Chain Monte Carlo (MCMC) method) which obtain a probability distribution in cases where its direct calculation would result in unfeasible computing times. A range of algorithms to sample the target distribution have been developed, each being more optimal depending of the studied problem. In the case of Organic Residue Analysis, FRUITS applies the commonly used Metropolis-Hastings algorithm in its “Gibbs sampling” variation. In this process, the posterior probability distribution is approximated by selecting a random value, evaluating whether it would belong to the distribution or not and then jumping into another one. When this approach is repeated a sufficient number of times, the retained values are considered to be a good approximation of the posterior distribution.

Model convergence can be defined as the degree to which a truly representative solution set has been generated (Ramsey, 2009). Not achieving it implies that, each time the model is run, its outcomes will be different because the MCMC algorithm has not generated a complete picture of the posterior distribution yet. Therefore, it is of outmost importance to validate that convergence has been achieved. In FRUITS, the implemented mechanism to assess convergence is to plot the α_i values resulting from all iterations (see Figure 16). When an asymptotic behaviour is observed, model convergence can be assumed (Fernandes *et al.* 2017).

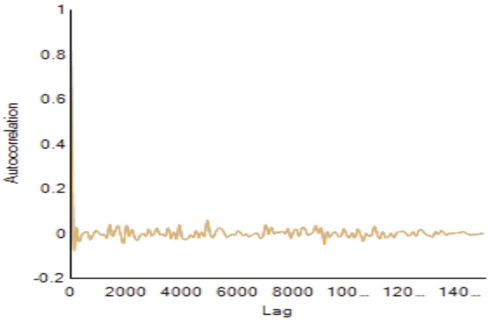


Figure 16: An example of a convergence test in FRUITS. In this case convergence is achieved after only a few iterations as the autocorrelation value quickly drops to around 0.

Therefore, the number of times the model is run (its iterations) has to be high enough to guarantee reproducibility. As a precaution, the first 5000 iterations are discarded (burn-in steps) so that results from a non-converging model are not passed on the final report. After that, models are usually run for extra 10000 iterations (Fernandes *et al.* 2017). If higher precision in the posterior distribution is needed, the number of iterations can be raised.

Once the posterior probabilities have been calculated and the Bayesian model completes its job, the data might be treated so that hypotheses and archaeological questions can be better evaluated. The following are the two most common data treatment processes used when working with source data and posterior distributions.

3.5.2 Data treatment: source combinations

As previously discussed, mixing models of all types are especially sensible to the amount and the choice of sources. Results will only contemplate the possible quantities of initially defined sources and, consequently, any modification will severely alter the results (Phillips *et al.* 2005). Therefore, any model must be built bearing in mind all the available information for each case study and the specific archaeological question to be answered. Given these limitations, all mixing models are ill-suited for general explorative analyses where the researcher does not know the range of products that could have been incorporated into the residue or their number is really high.

When the amount of sources is significant, they can be combined either a priori or a posteriori (Phillips *et al.* 2005) to provide results which better answer a specific archaeological question.

As a priori combination can be applied to sources which present a certain degree of isotopic similarity and usually following a logic relation, for example, the combination of dairy ruminant and adipose ruminant distributions into a general ruminant fat source (Fernandes *et al.* 2017). Alternatively, although freshwater fats and plant oils can present similar values, its combination would not be advised given that the interpretation of the posterior mixing estimates would be extremely difficult (Phillips *et al.* 2005). This can be particularly useful to reduce the amount of sources and produce clearer results but it also translates into increased isotopic variability and, therefore, greater uncertainties (Phillips and Gregg 2001, Phillips *et al.* 2005). This approach is common practice when producing reference data in organic residue analyses. The studied products are almost always groups of species separated by physiologic and environmental differences which justify significant shifts in its isotopic ratios: ruminant adipose fats, non-ruminant adipose fats, freshwater fish and marine fish are just some examples (ex: Dudd and Evershed 1998, Colonese *et al.* 2017b).

A posteriori combinations can be useful when aggregation is not mathematically advisable but still necessary to aid interpretation (Phillips *et al.* 2005). As an example, a study evaluating the importance of aquatic (both freshwater and marine) resources will not be able to generate an a priori group including both types given the significant offset in $\delta^{13}C$ values between marine and freshwater organisms. The increased isotopic variability resulting from such combination would be greater than the difference between other possible sources, thus producing extremely uncertain estimates which could prove useless to solve the archaeological question. Alternatively, freshwater and marine can be combined a posteriori. In this case, the model will provide density estimates for both sources and, afterwards, these will be added so that the new combined source presents higher quantity estimates than the previous sources individually. This approach has been successfully applied in paleodietary studies on human collagen (Newsome *et al.* 2004) but, so far, it has not been reported in models working with lipids from pottery organic residues. Nevertheless, the use of a posteriori combinations may carry some statistical issues. The act of aggregating two source posteriors results in a combined source posterior that will reflect two aggregated priors. Given that the uninformative prior is built so that each source is equiprobable, the combined source will now represent twice the weight of the remaining source priors (Stock *et al.* 2018). Therefore, the more a posteriori additions are practiced in the results from the mixing model, the more biased the model will become towards the new groups. One way to avoid this problem is to perform the necessary a posteriori additions so the result is an equally probable range of grouped sources (Stock *et al.* 2018). For example, a model with 6 sources which assumes each source will be 1/6th of the mixture can be combined in three new groups each containing two of the original sources so that each will then be assumed to contribute 1/3rd into the mixture. Another option could be to include a prior which reduced the expected contribution of those sources to be combined a posteriori so that, after the addition, the resulting groups become equally probable. Alternatively, were biases from a posteriori additions not avoided intentionally, these could be conceived as a prior and, therefore, they should be supported by archaeological data.

The process of calculating a posteriori source additions is not straightforward and also not programmed into FRUITS. Initially, this can be conceived as the sum of two statistical distributions. To execute it, the nature of the relationship between sources must be first taken into account given that the intersection of each pair of probabilities will be calculated. All posterior distributions are conditioned by the fact that a higher quantity in one source implies a lower quantity on the others. This is true because they are conceived as the components of a mixture expressed in percentages. Nevertheless, when combining two sources a posteriori, the resulting distributions have already taken into account this condition and, therefore, instead of calculating the intersection of two conditioned distributions:

$$P(A \cap B) = P(A) \cdot P(B|A) \qquad 5$$

The two can be considered independent from each other, and therefore:

$$P(A \cap B) = P(A) \cdot P(B) \qquad 6$$

In consequence, for each percentage point in the amount of the new combined source, their probability (P(C)) is equal to the summation of all the intersections of the probabilities of values A and B given that the addition of the values of A and B is equal to C.

$$P(C) = \sum_{\substack{i,j \\ P_i(A)+P_j(B)=C}} P_i(A) + P_j(B) \qquad 7$$

This is, in essence, the same procedure used when combining the probabilities of getting a certain result after rolling two dice. First, all the possible outcomes are taken into account. When rolling two dice, these are 36 :

1: first dice rolls a 1 and second dice rolls a 1,
2: first dice rolls a 1 and second dice rolls a 2, ...
36: first dice rolls a 6 and second dice rolls a 6

Nevertheless, in the case of the quantity of a certain product which might have been present in the residue, FRUTS contemplates 100 cases (one for each percentage from 1% to 100%), which results in 4950 possible outcomes where the addition of the two quantities is 100% or less:

1: first product is 1%, second product is 1%
2: first product is 1%, second product is 2%
...
200: first product is 30%, second product is 45%
...
4950: first product is 99%, second product is 1%

When this data is available, the intersected probabilities of all the possible outcomes are calculated. In the case of two dice:

Chance of rolling a 1 in the first dice = 1/6 and
chance of rolling a 1 in the second dice = 1/6. Therefore:
1/6×1/6=1/36

In consequence, the chance of obtaining a value of 2 when rolling two dice and adding the results is 1/36. When working with lipid sources, the probability for each source will usually be different, for example:

Chance the first product contributed 1% into the
mixture = 1/100 and chance the second product contributed
1% into the mixture= 1/7. Therefore: 1/100×1/7=1/700

Finally, the probabilities from all the outcomes with the same added values are all summated. In the case of the dice, there are four cases where the addition is 5: rolling 1 and 4, rolling 2 and 3, rolling 3 and 2, and rolling 4 and 1, each with a 1/36 chance of happening. In conclusion, the chance of obtaining a value of 5 when rolling two dice and adding the numbers is 4/36. In the case of lipid sources, the amount of cases to summate can be higher, for example, there will be 49 possible combinations where the two sources represent 50% of the mixture.

After performing this procedure for all possible percentage contributions to the mixture, the resulting distribution, as expected, will present higher quantities that the two initial ones. Given that executing this algorism for 4950 cases in each sample is severely time consuming, a script can be programmed to automate the task. The R function "combine_sources" (Stock *et al.* 2018) can be used to perform these calculations. Alternatively, to work with results from FRUITS, the aforementioned algorism has been programmed in a PHP script and inserted in a user-friendly webpage which can be accessed by anyone interested in performing such calculations. The source code of the PHP script can be found in appendix 1.

3.5.3 The Bayesian model applied in this research

3.5.3.1 Priors

When studying the best appropriate sources to analyse lipids from the north-western Mediterranean region, several factors must be taken into account:

1: Isotopic fatty acid values have been shown to vary across climatic regions and, therefore, not all available references in the literature would be comparable with data obtained from the Catalan seashore (Muckherjee *et al.* 2005).

2: To date, there are no compound specific isotopic values available from authentic modern reference fats sampled from animals raised in the region. Although research such as Tarifa (in preparation) is developing a reference database, the values are not yet available.

To overcome such difficulties, a range of reference datasets collected from neighbouring countries presenting more cold and wet or dry and arid climates have been taken into account. The data is mostly derived from the database prepared by Drieu (2017) with only minor modifications to account for recent additions. Reference values resulting from grazing on C4 plants and extremely enriched cases which do not correspond with the expected range of cases usually obtained in C3 grazing regions have not been taken into account. This approach provides the model with a range of variation which will encompass the specific expedted values of the Catalan coast.

In all cases, three sources were prepared: Dairy products, Ruminant adipose fats and Non-ruminant adipose fats. These have been chosen because of the following archaeological criteria:

1 Detection of animal bone remains across the studied archaeological sites and other contemporary settlements indicate the availability of animal protein including ruminant and non-ruminant species. Furthermore, fatty acids coherent with the degradation of animal fatty acid triacylglycerols have been detected in organic residues from pottery in the same region (Spiteri *et al.* 2016, Drieu 2017, Tarifa *et al.* 2019).

2 Although present in the archaeological record mostly as carbonised grains, pollen, phytolyths and charcoal, the inclusion of a plant source in the model would not be advisalbe unless biomarkers suggesting any significant input of plant matter are detected. This should not be mistaken as proof of the absence of plant residues in these pottery vessels.

3 Similarly to the case of plants, although some archaeological evidence such as seashells and minimal amount of fish bones in some sites suggest marine products could have been consumed, both freshwater and marine sources should not be included unless significant molecular biomarkers supporting its presence are detected. No organic residue analyses so far have reported marine molecular fingerprint in mediterranean prehistoric pottery.

4 A certain amount of wild animals were still being hunted at the onset of the Neolithic. This has been demonstrated by the presence of wild taxa amongst the animal bone assemblage. Nevertheless, its percentage is always low and the domestic-wild distinction might not be reflected in the isotopic values unless it corresponds with a significant change in diet. The most commonly detected wild species in the studied archaeological sites were: Capra pyrenaica, Sus scrofa, Oryctolagus coniculus and Cervus elaphus. On a case by case base, Capra pyrenaica and Sus scrofa, given that they are the wild counterparts of domestic animals, were integrated in the Ruminant and Non-ruminant sources respectively. The two remaining species present isotopic values significantly different from other cases. Leporids are highly hervibore non-rumiant species which a specific digestive system (cecotrphia) that results in significantly depleted δ^{13}C isotopic values (Taché and Craig 2015, Choy *et al.* 2016). Nevertheless, given that the only available reference values so far originate from Alaska and Canada and it is not a major species in the overall archaeological bone assemblage, it has not been considered as a source in the Bayesian mixing model. However, its possible presence has been considered on a case by case basis depending on the exact composition of the faunal assemblage in each site. The same approach has been taken for Cervus elaphus. Given that this species tends to plot in an area similar to that from other ruminant fats (Craig *et al.* 2012) and partially overlap with dairy products but it is only found in minor quantities in the archaeological record, it has not been included as a source but it has been taken into account in those sites in which it could have significantly impacted the measured isotopic values.

5 Studies on herd management and kill-off patterns (Saña and Navarrete 2016, Saña *et al.* 2015, Gillis *et al.* 2016, Spiteri *et al.* 2016) suggest that strategies which contemplated the acquisition of dairy products could have been in place. Moreover, previous organic residue analyses in the north-western Mediterranean have successfully detected dairy products in pottery vessels (Spiteri 2012, Drieu 2017, Tarifa 2019). In consequence, this product has also been included as a source.

Although the herd composition suggests a significant dominance of ruminant over non-ruminant species and the amount of meat and fat yielded by cattle is significantly higher than what can be obtained from pigs, no informative priors reflecting this reality were implemented. Alternatively, a posteriori additions practiced due to model constraints (see below) slightly biased the Bayesian mixing model towards ruminant adipose fats.

Furthermore, to validate the accuracy of the model, all the modern reference samples used to construct the source descriptors were run as unknowns. In this test, the model was considered to accurately predict the source when the product with the highest median corresponded with the nature of the sample. The results could be then studied to evaluate which modifications in the construction of the sources could lead to an improvement of the model accuracy.

The specific values used in each case are presented in appendix 2.

First model

In this model, all modern samples complying with the aforementioned conditions were included in the study. 75 dairy, 36 ruminant and 28 non-ruminant measurements were identified as follows (Table 7):

Model/Origin	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	71	1	0
Ruminant	4	32	0
NonRuminant	0	9	19

Table 7: Highest median returned by the model compared with its true origin in the fist model.

Therefore, almost all dairy products were correctly identified (71/75) and only misidentified as ruminant adipose fats. Ruminants were also majorly well identified (32/42) although a higher number of cases were wrongly classified as non-ruminant products. Finally, all non-ruminant samples were correctly assigned by the model. The results have been normalised (Table 8) for the sample size in order to obtain an accuracy estimate which can be compared with other models.

Normalised	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	0.99	0.01	0.00
Ruminant	0.11	0.89	0.00
NonRuminant	0	0.32	0.68

Table 8: Normalised results compared with true values in the first model

In conclusion, there is a significant amount of false non-ruminant positives. This is due to highly enriched ruminant values not being taken into account. One explanation might come from the fact that the reference database contains a lot of samples from northern European countries. Because these are overrepresented, ruminant values lower than -26‰ appear to be outside its ranges but inside the ranges in non-ruminants, thus causing the misidentification. To attempt to solve this issue, the number of data points across the ruminant value range should be equilibrated, which has been achieved by removing the UK reference samples. Therefore, the Ruminant source in the second model will be composed only of reference values form Palestine, Israel, Malta, Switzerland and Kenya.

Second model

The second model tested the same amount of unkown samples as model 1 with these results (Table 9):

Model/Origin	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	74	5	0
Ruminant	1	28	0
NonRuminant	0	9	19

Table 9: Highest median returned by the model compared with its true origin in the second model.

As presented, the amount of correctly identified dairy products has increased to 74/75, but it has fallen in Ruminants (28/42) without affecting its accuracy. Finally, all non-ruminant samples were again correctly assigned. The normalised results (Table 10) will help gain a fuller picture.

Normalised	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	0.94	0.06	0.00
Ruminant	0.03	0.97	0.00
NonRuminant	0.00	0.32	0.68

Table 10: Normalised results compared with true values in the second model

Although the trueRuminant value increased from 0.89 to 0.97, the problem with the enriched ruminant samples was not fully solved yet, as there was still a 0.32 probability of a false non-ruminant positive. Moreover, the ratio of Non-ruminant accuracy was still in an uncomfortable 0.68. To reduce the amount of TrueRuminant identified as NonRuminant and increase the reverse, the ruminant source was split in two to take into account its huge variety of absolute values. Now, because there was no need to exclude UK samples, they were again included in Ruminant A source, which comprised 32 values from Switzerland, Palestine, UK, Malta and Kenya. The Ruminant B group included 10 values only from Kenya, Malta and Israel (Table 11).

Modelled/Origin	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	69	4	0
Ruminant	5	35	1
NonRuminant	1	3	18

Table 11: Highest median returned by the model compared with its true origin in the third model.

The third model tested the same amount of unknown samples as model 1 and 2. Given that Ruminant A and Ruminant B were essentially the same products, its results were added into the TrueRuminant column so that the data could be comparable with previous models:

Normalised	TrueDairy	TrueRuminant	TrueNonRuminant
Dairy	0.95	0.05	0.00
Ruminant	0.12	0.85	0.02
NonRuminant	0.05	0.14	0.82

Table 12: Normalised results compared with true values in the third model

In this case, the amount of correctly identified dairy products decreased to 69/75 while the ruminant adipose fats present the highest score so far (35/42). For non-ruminants, one sample was misidientified as a ruminant (18/19). Therefore it seemed that the improvement in the separation between ruminant and non-ruminant might have come at the expense of dairy products. For a better understanding, the normalised values were calculated (Table 12).This time, the amount of Ruminants misidentified as NonRuminants was reduced by half and the amount of correctly identified non-ruminants increased up to 0.82. Nevertheless, these results were achieved at the expense of increasing overall uncertainty given that now there was a possibility higher than 0 that a dairy product was misclassified as a non-ruminant, and a non-ruminant was classified as ruminant. Alternatively, correct identifications of NonRuminants also increased but there the model seemed to be slightly more skewed towards producing false dairy product identification. Nevertheless, this factor could be assessed by checking Δ^{13}C values.

In conclusion, to evaluate which model might be more optimal, its overall accuracy, the mean of the errors and its standard deviation were calculated.

Model	Accuracy	Mean error	SD error
1st	0.897	0.073	0.117
2nd	0.890	0.068	0.115
3rd	0.897	0.063	0.051

Table 13: Accuracy, Mean error and the Standard deviation of the error for the three models

As presented, the third model shows the same accuracy as the first one but has a lower mean error and more than half the standard deviation of the error (Table 13). This indicates that it is not only more accurate but also more precise and generally less biased. In conclusion, this is the model which has been used. Such a choice implies that, in order to correctly asses the possible quantity of Ruminant adipose fats in the mixture, a posteriori addition of the Ruminant A and Ruminant B sources will have to be performed. This will create a bias in the model, which, as previously discussed, does not generate any conflict with the available archaeological data.

3.5.3.2 Likelihood

Source descriptors are defined by repeated measurements of the values in modern reference samples. The mean, the STE_{mean} and the variance-covariance matrix calculated from these measurements are introduced into the model. All data in this stage has been already corrected for the Suess effect and, therefore, no offset parameter (T_{ik}) is used. Finally, given that the amount of palmitic and stearic acids in triacylglycerols is not the same across different species; its concentration parameters (the mean, the STEmean and the variance-covariance matrix) are also feeded to the model. Furthermore, as indicated by Fernandes *et al.* (2017), this is a non-weighted model given that it is assumed that the source of carbon for a particular fatty acid extracted from the ceramic matrix can only be the same fatty acid found in the lipid sources, excluding taphonomic effects.

In this case, the reference data used has been the following one (Table 14):

Source	**Dairy**		**Ruminant A**		**Ruminant B**		**Non-Ruminant**	
Fatty acid	δ^{13}C18:0‰	δ^{13}C16:0‰	δ^{13}C18:0‰	δ^{13}C16:0‰	δ^{13}C18:0‰	δ^{13}C16:0‰	δ^{13}C18:0‰	δ^{13}C16:0‰
Mean	-32.4208	-27.7508	-31.5675	-29.8052	-27.0500	-26.2740	-25.5711	-26.5632
STEmean	0.271	0.226	0.153	0.145	0.237	0.302	0.327	0.271
Concentr.	18.4	34.6	28.5	24.2	28.5	24.2	11.5	24.1
St. Dev.	8.4	7.2	6.5	5	6.5	5	2.4	2.1

Table 14: Reference data used to study archaeological data.

As previously stated, the values used to calculate the expected mean and its standard error of the mean are presented in appendix 2. Regarding concentration data, the information provided in Fernandes *et al.* (2017) has been used in this case as well. Variance-covariance tables were calculated for each source by studying all pairs of possible combination of variables ($\delta^{13}C16:0$ and $\delta^{13}C18:0$). After multiplication of the variances of every possible combination of isotopic values, the values were summated and divided by the number of reference samples available in that source (n) to result in Table 15:

	$\delta^{13}C_{16:0}$	$\delta^{13}C_{18:0}$
$\delta^{13}C_{16:0}$	$\frac{\sum_{i=1}^{n}(x_{i\,16}-\overline{x}_{16})^2}{n}$	$\frac{\sum_{i=1}^{n}[(x_{i\,16}-\overline{x}_{16})\times(x_{i\,18}-\overline{x}_{18})]}{n}$
$\delta^{13}C_{18:0}$	$\frac{\sum_{i=1}^{n}[(x_{i\,18}-\overline{x}_{18})\times(x_{i\,16}-\overline{x}_{16})]}{n}$	$\frac{\sum_{i=1}^{n}(x_{i\,18}-\overline{x}_{18})^2}{n}$

Table 15: Formulae used to calculate the variation-covariation tables.

In consequence, the specific variation-covariation tables (Table 16) were:

Source	**Dairy**		**Ruminant A**		**Ruminant B**		**Non-Ruminant**	
Isotope	$\delta^{13}C16:0‰$	$\delta^{13}C18:0‰$	$\delta^{13}C16:0‰$	$\delta^{13}C18:0‰$	$\delta^{13}C16:0‰$	$\delta^{13}C18:0‰$	$\delta^{13}C16:0‰$	$\delta^{13}C18:0‰$
$\delta^{13}C16:0‰$	3.64	3.71	0.65	0.36	0.51	0.32	1.35	1.12
$\delta^{13}C18:0‰$	3.71	5.22	0.36	0.73	0.32	0.82	1.12	1.33

Table 16: Variation-covariation tables used to study archaeological data.

Regarding the archaeological data, the uncertainty associated with each isotopic measurement was set at the common value for measurement reproducibility (0.3‰)(Fernandes *et al.* 2017) but the minimum uncertainty, which is a value associated with each parameter of the model, was set at double the previous figure (0.6).

3.5.3.3 Posteriors

As previously described, the recommended 5000 burn-in steps were performed and, afterwards, the model run for 25000 iterations. Although, this is higher than recommended figure, some cases benefited from additional calculations which increased the precision of the results.

After the "a posteriori" additions, results for selected samples were averaged to represent specific phenomena. Following this approach the mean results for each archaeological site and the mean abundances in specific archaeological types were calculated. In this case, the summation of the probabilities of each quantity percentage was divided by the number of samples being averaged.

$$\mu = \frac{\sum_{i=1}^{n} P(x_i)}{n} \qquad 8$$

The obtained mean would, thus, become a representation of the importance of a specific product as a ceramic organic residue in a specific archaeological site or pottery type, which can be used to study differences between regions, periods or pottery traditions (see Figure 17). When comparing such phenomena, differences are more evidently represented when the probability distributions are subtracted. The remaining is a curve indicating the probability a certain source was more abundant in one (for example) pottery type than another.

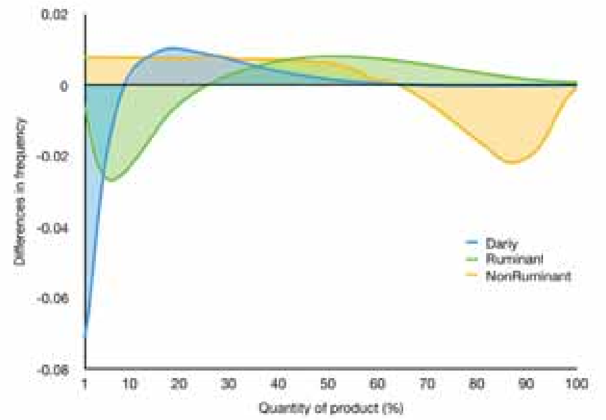


Figure 17: Example of the type of graph obatined as a result of subtracting the density probability distributions retured by FRUITS. Negative or positive values in the x axis reflect a higher probability that one of the compared groups presented a given quantity (y axis) of a given product (each curve).

In conclusion, working with Bayesian models provides a range of advancements which aids in the interpretation of the archaeological data. It offers the possibility of better managing the accumulated uncertainties of archaeological and analytical work as well as constraining the space for subjective interpretations given that hypotheses are directly linked with the data and a probability estimate. Moreover, the inclusion of a wide range of factors helps the researcher arrive at more nunaced interpretations.

Nevertheless, Bayesian models also pose several limitations. When working with such complex mathematical models it is very easy to fall into a "black box" situation where the researchers are completely unaware of the mechanisms that are transforming their data. In cases such as this one, where the archaeological question must be well tailored into the model, one-size-fits-all approaches will almost certainly present unexpected biases which could affect the overall interpretation and could fool the researcher into a false conclusion. To correctly work with such powerful statistical tools, a significant learning curve must be surpassed.

In essence, mixing models are usually as good as their source data. A deep understanding of which elements might have been mixed together and a good knowledge of the archaeological data supporting this hypothesis is of outmost importance not to fall in circular arguments. Moreover, a good understanding of what results could be expected and its meaning and further implications is also vital not to reach conclusions which could have been easily reached without the need of such mathematical complications.

Bayesian models, a tool capable of managing uncertainty and providing a framework to rationalize usually extremely difficult social questions might be of great value to a discipline which is surrounded by degraded, destroyed or fragmented data. But, as aphorism in statistic says “All models are wrong, but some are useful”, attributed to Box (1976). Modelling won’t solve all the problems. The quality of the data used is the key to obtaining results which help better understand the mechanisms in motion behind past societies.

Results and archaeological contexts

4 The Barcelona Plain

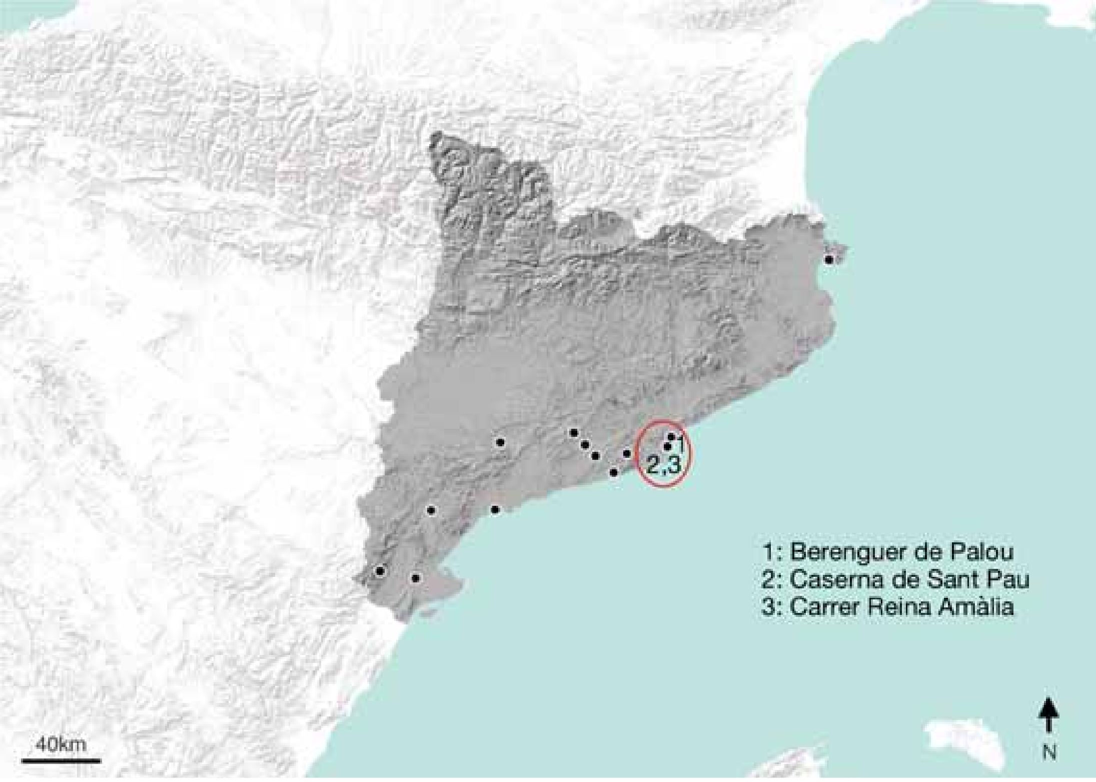


Figure 18: Map of the studied site in the Barcelona plain.

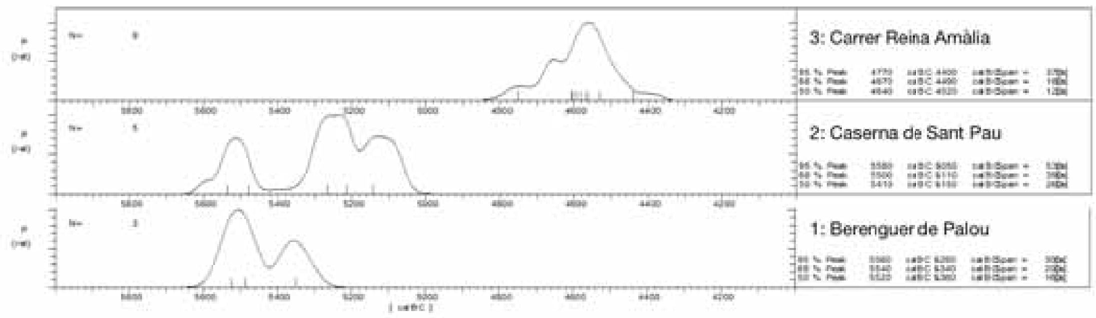


Figure 19: Calibrated ^{14}C dates from Early Neolithic contexts in the Barcelona Plain. Dates from Berenguer de Palou were performed on seashells and calibrated with the Marine IntCal04 curve. Software: CalPal v.15

4.1 Berenguer de Palou

4.1.1 Introduction

This archaeological site was excavated between the end of 2017 and the year 2018 as contract work prepared the development of a housing block in Barcelona. It is located in the Sant Andreu quarter, about 3km from the seashore and around 20m above the sea level.

4.1.2 Archaeological features relevant in this study

After three trenches were practiced to evaluate the possible existence of archaeological remains, the site was excavated extensively and a set of 14 Early Neolithic negative structures interpreted as silos were detected and excavated (Pereira 2019). The debris inside included lithic tools (some of them tentatively identified as jasper) and pottery decorated following the cardial technique but, aside from a set of seashells, it did not contain animal bone remains. The flotation of the sediment in the silos did not reveal any significant presence of charcoal or charred seeds which could help better understand the economy and chronology of the site. Given the absence of other valid materials, seashells from the silos 413, 450 and 210 were ^{14}C dated from the second half of the VIth millenia, which corresponds with the Early Neolithic. The study of the marine remains recovered from the site has so far reported the existence of perforated seashells which could have been used as jewellery.

4.1.3 Archaeological materials

The initial reports of the study of the ceramic assemblage indicates the existence of a set of sherds coherent with the technological choices already known in the Barcelona plain (mixed firing atmospheres and minimally treated surfaces). The cardial imprints coexist with linear appliqués and handles, which compose the expected range of chronocultural markers usually recovered from Early Neolithic sites in the north-east of the Iberian Peninsula. Moreover, it has been hypothesised that, given the apparent similarity between clays from this site and Caserna de Sant Pau, that vessels could have been produced in the Barcelona Plain (Gómez 2019).

4.1.4 Sampling and analytical techniques

Two wall sherds recovered during flotation of stratigraphic units 458 and 438 were sampled to evaluate the extent of organic matter degradation in the site. After removal of the surface, around 1g of sample was drilled and extracted following the acidified methanol procedure in the ICTA laboratories.

4.1.5 Results and discussion

LAB ID	UE	Shape	TLE	Interpretation
BDP1	458	Wall	3.76	Non significant residue
BDP2	438	Wall	2.05	Non significant residue

Table 17: Samples and total lipid extracts ($\mu g \cdot g^{-1}$) from Berenguer de Palou after GC-FID analyses.

Analysis through Gas Chromatography did not reveal the presence of any significant amounts of lipids which could be attributed to an archaeological residue (Table 17). Moreover, previous analytical work in the same area on grave goods from a IVth millennia communal grave was not able to recover significant organic residues either (Molina 2015b).

4.1.6 Conclusion

In consequence, further work will have to be carried out in order to fully understand wether the absence of archaeologically significant residues could be due to aggressive degradation phenomena or a different pattern of use compared with other Early Neolithic sites in the Barcelona plain (Caserna de Sant Pau and Carrer Reina Amàlia).

4.2 Caserna de Sant Pau

4.2.1 Introduction

After several development works motivated by the Olympic games between 1988 and 1992, the demolition of the old Civil Guard barracks next to the Sant Paul's Monastery in Barcelona revealed significant prehistoric remains (Granados *et al.* 1993). The site, 5 meters above the sea level and less than 1km away from the modern beach, was also really close to the ancient seashore (Julià and Riera 2010, 2012) and to the northern slope of the Montjuïc mountain. As expected from an urban excavation, Roman, Bronze Age and Neolithic occupations were detected across the whole stratigraphic sequence. The fieldwork was performed by Robert Farré and coordinated by the archaeological unit in the Barcelona History Museum and the excavation's preliminary results were presented in several publications (Granados *et al.* 1993; Anfruns *et al.* 1995, Laorden *et al.* 1993). Later, the site's potential motivated a comprehensive research project to improve the archaeological knowledge available on the first Neolithic settlers in the Barcelona plain. This (Molist 2006), culminated with the publication of a series of new results covering the whole prehistoric chronological sequence at Caserna de Sant Pau (Molist *et al.* 2008; Molist 2009, Gómez *et al.* 2013, Gómez and Molist 2017).

Further contract archaeological work has consistently detected Neolithic features in the streets next to the Sant Pau Monastery. This includes, amongst others, excavations in Reina Amàlia Street nº 31-33 (included in this study), Reina Amàlia nº 38, C/Nou de la Rambla or C/Beates 2 and more (see Molist *et al.* 2016). As suggested by ^{14}C dates and the nature of the human occupations, it seems that a significantly wide and multi-period Neolithic open air settlement is located underneath the modern Raval quarter (Molist *et al.* 2016). More recently, further excavations in the Sagrera quarter have also revealed the existence of other Early Neolithic prehistoric remains (including the previous studied site, Berenguer de Palou). The distance between these two clusters suggests the existence of two different settlements.

4.2.2 Archaeological features relevant in this study

Caserna de Sant Pau presents five sedimentary layers which are the result of a series of successive human occupations:

Level I: featured mainly silt and clay with no archaeological remains.

Level II: between 0.3 to 0.5m deep, contained materials from the early and Late Bronze Age.

Level III: between 0.5 to 1.3m deep, was composed of sand and gravel with a clay matrix. It was also deprived of any archaeological materials.

Level IV: was around 0.6m deep and 800m^2 wide and contained silty and clayey dark sediment rich in organic matter. The Neolithic remains were found in this level.

Level V: mainly limestone blocks and no archaeological remains suggested a purely geological origin which marks the end of the excavation. Several negative structures were dug in this layer.

Excluding more recent archaeological evidences, the Neolithic features were a series of inhumations, hearths, silos and possible occupation layers. The 24 burials, a testimony of possibly the oldest Neolithic necropolis in the Barcelona plain (Molist *et al.* 2008), were distributed in two groups: 9 graves in the north east and 15 graves in the south-est. The two clusters were roughly 10 to 15m away. This necropolis has been linked to occupation levels containing different types of combustion structures (flat hearts and buried hearts) at different depths. Further spatial analytical work (Vicente *et al.* 2014) was able to detect the existence of three occupation moments: the Cardial Early Neolithic, and two postcardial Neolithic phases. The oldest phase would also incorporate the presence of small negative structures interpreted as postholes and a set of nine silos (see Table 18) placed below stratigraphic unit IV.

Structure number	Depth	Maximum diameter	Plan	Walls	Base	Sampled
1	1.65	1.27	Circular	Unkown	Concave	Yes
2	1.69	1.2	Circular	Concaves	Unkown	Yes
3	0.79	0.58	Circular	Straight	Flat	No
4	1.14	1.2	Circular	Straight	Flat	Yes
5	1.55	0.73	Circular	Concaves	Unkown	Yes
9	1	0.9	Circular	Unkown	Unkown	Yes
10	1.6	1.1	Circular	Unkown	Unkown	Yes
11	0.85	0.75	Circular	Unkown	Unkown	No
14	0	0.9	Circular	Unkown	Unkown	Yes
15	Unkown	Unkown	Circular	Unkown	Unkown	No

Table 18: Characteristics of the 9 Early Neolithic silos in Caserna de Sant Pau. Adapted from Molist *et al.* 2008

The silos, all dug in level V (limestone from geological origin), contained sherds with Cardial decorations and were distributed in two clusters. The first, in the site's central area, contained more recent ^{14}C dates while the second (silos 9 and 10), in the site's south-east corner, contained older ^{14}C dates (see Table 19). The structures themselves were in a good preservation state, including some complete in some cases. They had a circular rim, troncoconic profiles, either flat or rounded bottoms, and were roughly 1m deep and less than 1m wide, which corresponds with the expected parameters in the region (Molist *et al.* 2016).

Labcode	Silo	Material	Date	Calibrated at 2σ
Beta 236174	1	animal bone	6290±50	5372-5076
Beta 236175	2	animal bone	6250±40	5316-5070
Beta 407494	2	charred seed	6200±30	5289-5051
Beta 407495	9	charred seed	6590±30	5615-5482
Beta 407496	10	charred seed	6510±30	5534-5380

Table 19: ^{14}C dates from the Early Neolithic layers hitherto published in Caserna de Sant Pau. Reproduced from Gómez and Molist 2017.

As suggested by the ^{14}C dates (see Table 19), silos 9 and 10 would have been filled with domestic seeds significantly older than the animal bones found in silos 1 and 2. This evidence points at Caserna de Sant Pau as one of the oldest Cardial Neolithic sites in the Catalan seashore and suggests silos were dug, maintained and abandoned progressively during a long period of time. Taking their spatial distribution into account, it should not be ruled out that the remaining non ^{14}C dated silos were chronologically similar to silos 1 and 2. This recursive use of the space could suggest that the population linked with said storage features permanently inhabited the Barcelona plain (Gómez and Molist 2017).

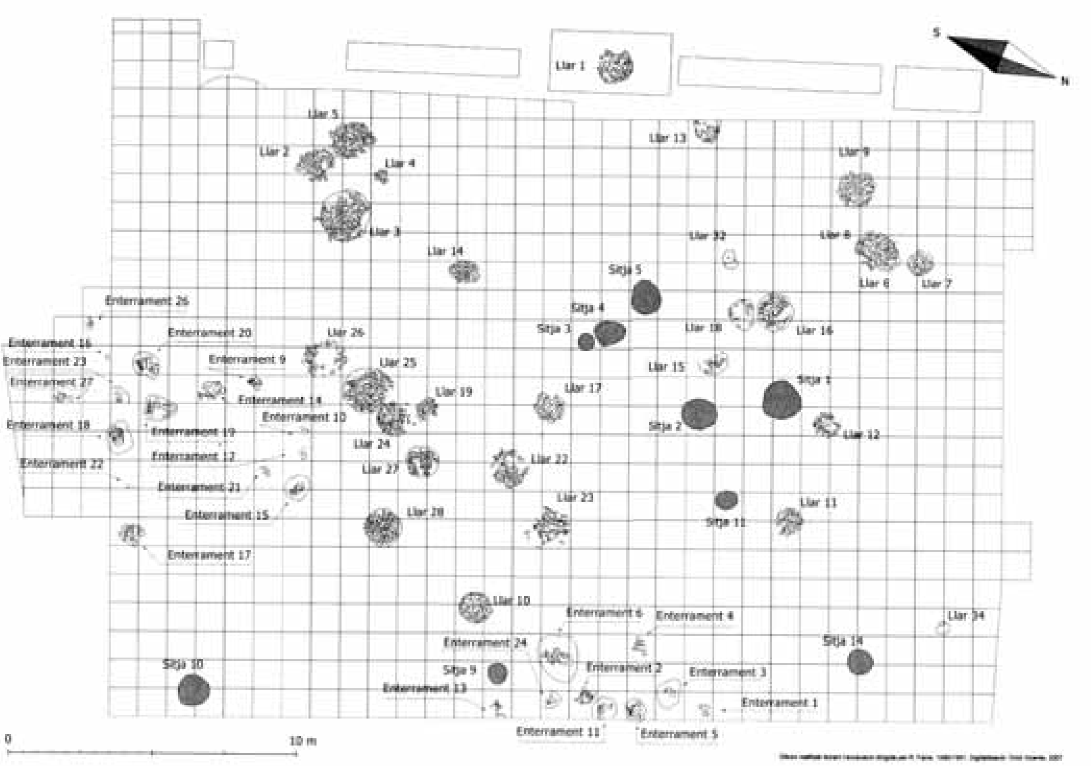


Figure 20: Plan including all Cardial and Postcardial Neolithic features from Caserna de Sant Pau. Reproduced from Molist *et al.* 2008.

4.2.3 Archaeological materials

4.2.3.1 Pottery remains

Regarding pottery, these negative structures included 724 ceramic fragments (Gómez *et al.* 2008). Overall, the preservation of the sherds in the silos could be defined by a low erosion and concretion index (Molist *et al.* 2009) and a fragmentation index around 9%. Following the analytical description developed by Dedet-Py, the assemblage presented rims suggesting mainly open shapes with rectilinear profiles or rounded rims (type D and E, 30% and 7%) followed by straight shapes (F, 27%) and closed shapes (H, 23%). Some rims could not be securely identified (13%). Other significant features in the assemblage were the low percentage of flat bases (2%) and the presence of ribbon (15%), over-elevated (2%) and tubular (3%) type handles. While a significant amount of vessels could have been middle sized (45%) big vases were also present (2%). Regarding shapes, middle to large sized bowls and big deep or middle neckless vessels were accompanied by the characteristic globular closed and necked jars from this period (Molist *et al.* 2009). The clays used presented a significant quantity of middle sized quartz, biotite and a variable presence of feldspars. After Gómez *et al.* (2008), the geological data from the Barcelona plain suggests the pottery production in Caserna de Sant Pau would have used local clays. Moreover, the pottery would have been fired oxidising or mixed atmospheres and internal and external surfaces would have been mainly smoothed. These are expected technological characteristics for Early Neolithic pottery in the region.

The decorative techniques in Caserna de Sant Pau ranged from the most common cardial motives and plain or impressed appliqués with a variety of tools to a set of three sherds whose decoration has been temptatively attributed to a *sillon d'impresion* technique by Oms (2014), although the full characterisation of this horizon in the Iberian Peninsula is under development[3]. The cardial technique was present in around 87% of the decorated assemblage (Molist *et al.* 2009) and was performed with seashells of various sizes from 1.9 to 3.6cm wide which were apparently not coherent with the toothed seashells found in the silos (Breu 2013). Its use involved placing the exterior of the shell or its natix in vertical or inclinated positions. A experimental comprehensive study of the pottery assemblage in the silos detected that different types of gestures were often used to create the same decorative motive (Gómez *et al.* 2014a). Other decorative techniques are appliqués and incisions made with a variety of tools (see Table 20).

Cardium	**Finger**	**Plain appliquée**	**Impressed appliquée**	***Sillon d'impresion***	**Incision**	**Other impression**
70	1	15	3	3	5	2

Table 20: Amount of fragments presenting each decorative technique in the Early Neolithic silos in Caserna de Sant Pau (Breu 2014).

In Caserna de Sant Pau, similarly to other Early Neolithic sites, Cardial wares would have been performed in bands which might include hanging and/or limiting motives filled with linear or diagonal (including "cereal spike") compositions. The ones showing diagonal components tended to be in sherds thicker than 10mm. Alternatively, sherds thiner than 10mm only presented linear motives. Regarding the structure of the decoration itself, when compared to other nearby Cardial sites (Can Soldevila IV and Can Banús), the geometric shapes used show a strong dominance of the straight lines. The orientation of these lines would have been more variable and the rule followed to spread the geometric shape

---

[3] For example with semminars such as "Contextualitzando la cerámica impressa: Horizontes culturales en la Península Ibérica, celebrated on March the 29th and 30st of 2019.

across the vessel surface would present the highest variability with the lowest dominance of any particular rule. In conclusion, it would have been necessary to follow certain rules in order to replicate the Cardial decorative stile found in Caserna de Sant Pau del Camp. Nevertheless, although the detected degree of variability would not compatible with a set of strictly imposed rules, it would not be inconsistent with the existence of a certain degree of artistic freedom. Furthermore, analysis of the microspaces between imprints, shown to correlate with individual motor patterns, do not support a scenario where different producers would have developed conspicuous sub-styles within the cardial norm. Alternatively, it can not be rejected that different potters independently choose from a pool of shared interchangeable motives (Breu 2014).

4.2.3.2 Lithic remains

Following Bofill *et al.* (2008), at least 66 macrolithic pieces were found across stratigraphic layers at Caserna de Sant Pau. Regarding lithic tools, the site would present a diversity of knapping methods and techniques in the lithic production and in the retouched tool kit. Moreover, raw materials were managed differently according to the desired product and the use it would be put to. This diversity would not only be a response to the different characteristics of the raw materials, but also to the techno-economic requirements, the activities/needs of the settlement, and the chronological and cultural framework. This suggests that mainly different productive processes and activities were carried out at the Neolithic settlement of Sant Pau del Camp (Borrell 2008, Borrell and Gibaja 2012, Borrell and Molist 2012). In this matter, Caserna de Sant Pau fits within the general parameters of the Early Neolithic lithic assemblages: (1) heat treatment of some raw materials associated with the extraction of blades/bladelets; (2) the possible use of pressure as a knapping technique, (3) the semi-surrounding exploitation of small blade/bladelet cores, (4) a greater diversity of the provenance of silicious rocks in later levels and (5) a greater importance of blade production and certain types of retouched implements in the Epicardial and Postcardial Neolithic (Borrell 2008, Borrell and Gibaja 2012, Borrell and Molist 2012). Finally, use-wear analyses on pieces from the postcardial period suggest lithic tools could have had a significant range of uses in a wide variety of tool types. Although the main activity detected was linked to the harvest and transformation of cereals, skin scraping and wear related to contact with inorganic materials (such as pottery) was also present. Although no significant amounts of traces for the processing of animal products were identified, the authors do not rule out the posibility that this activity was also performed in the site (Gibaja 2008). In absence of use-wear data from the VIth milennia, this study helps complete the picture of well stablished farmer-shepherd human groups in the Barcelona Plain.

4.2.3.3 Animal remains

Regarding animal bones (Colominas *et al.* 2008), these were extremely rare in the Cardial silos. Only 25 remains were recovered and only 14 were assigned a species: *Bos taurus* (NR=9, 64%), *Ovis aries/Capra hircus* (NR=1, 7%) and *Sus domesticus* (NR=4, 29%). To provide some perspective, 2256 bones from at least 11 different species were studied across all Neolithic layers. These belonged to *Sus domesticus* (NR=91), *Bos taurus* (NR=64), *Capra sp*. (NR=12), and wild animals such as *Sus scrofa* (N=18), *Capreolus capreolus* (NR=1), *Cervus elaphus* (NR=4), *Capra pyrenaica* (NR=4), *Vulpes vulpes* (NR=5), *Meles meles* (NR=1), *Oryctolagus sp.* (NR=2) and *Canis familiaris* (NR=6).

At the genus level, 32% of the remains belonged to ovicaprines, 17% belonged to bovids and 13% belonged to suids. The remaining 36,5% of the assemblage could not be identified and 1.5% belonged to other wild animals (see table 3 in Colominas *et al.* 2008). In terms of the digestive system, remains in the silos were 71% ruminant and 29% non-ruminant and remains across the site were 83% ruminant, 17% non-ruminant and 0.1% cecotrophs. Thermal alterations in the bone and cutting marks suggesting the animals had been included in the human diet were present across several species in the site.

Recent work by Navarrete *et al.* (2017) studied the $\delta^{13}C$ and $\delta^{15}N$ bulk collagen values in suid bones from Caserna de Sant Pau. When results were compared to herbivores, similar $\delta^{13}C$ values validated that suids could have consumed C3 plants or C3 fed animal products. Regarding $\delta^{15}N$ values, pigs were on average 1.4% enriched. This suggested it should not be ruled out that a relatively higher proportion of animal sources contributed to their dietary proteins. The researchers indicated that this feeding practice could be associated with home-based management systems where pigs would have been enclosed or relatively free to forage only within the settlement, thus increasing the possibilities of ingesting animal foodstuffs.

Regarding marine products, around 38 undetermined ictiofaunal remains were reported by Colominas *et al.* (2008). More recent research has identified a sea urchin thorn and *Sparidae* teeth (Gómez and Molist 2016), but the majority of the marine remains is composed by 3951 seashells or seashell fragments. In this case, it has to be taken into account that the site's placement in the seashore makes is possible that seashell accumulations were due to fluvial, lacustre and marine dynamics. Therefore, it should not be ruled out that a significant amount of the seashell assemblage could have been generated without human involvement. Neverhteless, the distribution pattern of these remains is not a consistent gradient from the coast to inland, but a bimodal distribution around the postcardial structures. This phenomenon suggests that the involvement of human societies in the disposition of the seashells should not be ruled out (Nadal and Estrada 2008).

Regarding the possible consumption of these organisms, a significant input of marine products in the human diet is not coherent with the significant erosion detected in the majority of the seashells, and specially for *Glycymeris sp*. (Lloveras *et al.* 2014). Nevertheless, minimal erosion on pieces from *Patella sp/caerulea* implicate the consumption of this species must not be ruled out.

In detail, the silos containing cardial pottery contained only small amounts of seashells. Only in silo 2 two seashells from *Patella* sp. suggest some degree of consumption of marine foods could be linked with the oldest Neolithic phase in the site. Table 21, reproduced from Nadal and Estrada (2008) reports the total 27 remains recovered. Other silos from the same period (S1, S4, S9 and S10) did not apparently contain any marine products.

Species	**S2**	**S5**	**S11**	**S14**	**S15**
Glycymeris sp/*glycymeris*	6	6	1	3	7
Spondylus gaederopus	0	0	0	0	1
Acanthocardia tuberculata	1	0	0	0	0
Patella sp	2	0	0	0	0
Fish bones*	no	yes	no	no	no
Total	9	6	1	3	8

Table 21: Table presenting the amount of marine aimal remains from Early Neolithic silos (S) in Caserna de Sant Pau.Adapted from Nadal and Estrada 2008. *According to Gómez and Molist 2017.

4.2.3.4 Plant remains

Soil samples were collected from the Neolithic structures in the site and analysed by flotation of the sediment and laboratory identification of the recovered species. A total of 238 remains from 13 different taxa were found across the site. More specifically, 97 remains were found in the studied silos (1, 2, 4, 5, 9 and 10). They were 85 fragments of cerealia (either *Hordeum* or *Triticum*), 17 of which could be further determined to be *Hordeum vulgare/nudum* (n=11) or *Triticum aestivum/durum/diccocum* (n=6). Additionaly 1 seed of *Pisum sativum* was found in silo 2. Ruderal plants (*Chenopodium album* n=1 and *Labiatae* n=2) were also present in the assemblage. Moreover, 6 rachis possibly from *Triticum diccocum* could indicate cereals were cleaned by the same human group (Buxó and Canal 2008).

Although no studies of charcoal remains from the silos have been published to date, analyses on data from other phases in the site suggests the presence of a varied landscape with mediterreanean and submediterranean trees and shrubs. It seems that fuel necessities in posterior periods would have been mainly covered by tree wood (70,11%) although some shrubs would have been also used (Mensua and Piqué 2008).

4.2.4 Sampling and analytical techniques

In summary, the data recovered from the Early Neolithic silos in Caserna de Sant Pau suggests the presence of groups of farmers and pastoralists settled in the marine coastline since the start of the first half of the VIth millennia before the common era. Studies on the provenance of lithic and pottery tools indicate they could have been managing nearby resources (jasper and local clays) while transforming a marshy environment surrounded mainly by forests but also some open areas with shrubs and other species. Marine products were incorporated into the labour processes in the settlement and it has not been ruled out that some seashells were consumed. Barely, wheat, ruminant and a significant proportion of non ruminant animals were raised in the site to feed the inhabitants and the recovered pottery shapes (bowls, globular and necked shapes) are well suited for food preparation. Organic residue analyses, therefore, offers the opportunity to gain new knowledge on the culinary strategies practiced in the Barcelona plain and evaluate wether pottery had become a widely used tool or, on the contrary, some specific products were prepared without it.

To this end, 24 pottery fragments from silos 1, 2, 4, 5, 9 and 10 were sampled to evaluate the possible existence of lipid residues. Furthermore, sediment from a burial and a hearth in the postcardial chronological phase were selected to assess the possible presence of lipids in the soil.

Lab id	Silo	Square	Cut	Id	Shape	Firing	Rim Decoration	Plastic decoration	Weight (mg)
ABB1	1	H-I 11-12	XXVII to XXXI	47	Rim	Oxidised	Incised	Handle	100
ABB2	1	H-I 11-12	XXVII to XXXI	41	Rim	Mixed	Overelevated	None	100
ABB3	1	H-I 11-12	XXVII to XXXI	49	Rim	Mixed	Overelevated	None	100
ABB4	2	K 11-12	XXVII to XXXII	15	Rim	Oxidised	Incised	appliquée	100
ABB5	2	K 11-12	XXVII to XXXII	20	Rim	Mixed	None	appliquée	100
ABB6	2	K 11-12	XXVII to XXXII	23	Rim	Mixed	None	None	100
CSP10	14	E3	XIII	no id	Wall	Mixed	None	None	102
CSP11	9	S2	XXII-XXXII	no id	Wall	Mixed	None	None	102
CSP12	10	βγ/11-12	XXIII-XLII	no id	Wall	Oxidised	None	None	97
CSP13	10	βγ/11-12	XXIII-XLII	no id	Wall	Oxidised	None	None	101
CSP7	1	I12	XXII	no id	Wall	Oxidised	None	None	100
CSP8	1	HI/11-12	XXVII-XXXI	no id	Wall	Mixed	None	None	97
CSP9	1	I11	XIX	no id	Wall	Mixed	None	None	99
CSPA	2	K12	XIX	no id	Wall	Oxidised	None	None	105
CSPB	2	K11	XVI	no id	Wall	Mixed	None	None	100
CSPC	2	K12	XXI	no id	Wall	Mixed	None	None	112
CSPD	4	N14	XXVII	no id	Wall	Oxidised	None	None	103
CSPE	2	K11-12	XXVII-XXXIII	no id	Wall	Oxidised	None	None	113
CSPF	5	M15	XIV	no id	Wall	Mixed	None	None	101
CSPG	2	K11	XXV	no id	Wall	Mixed	None	None	126
CSPH	4	N14	XXVIII	no id	Wall	Mixed	None	None	105
CSPI	4	N14	XXVII	no id	Wall	Mixed	None	None	100
CSPJ	5	M15	XXIV-XXX	no id	Wall	Mixed	None	None	100
CSPK	2	K11	XXI	no id	Wall	Mixed	None	None	103

Table 22: Analysed pottery fragments from Caserna de Sant Pau

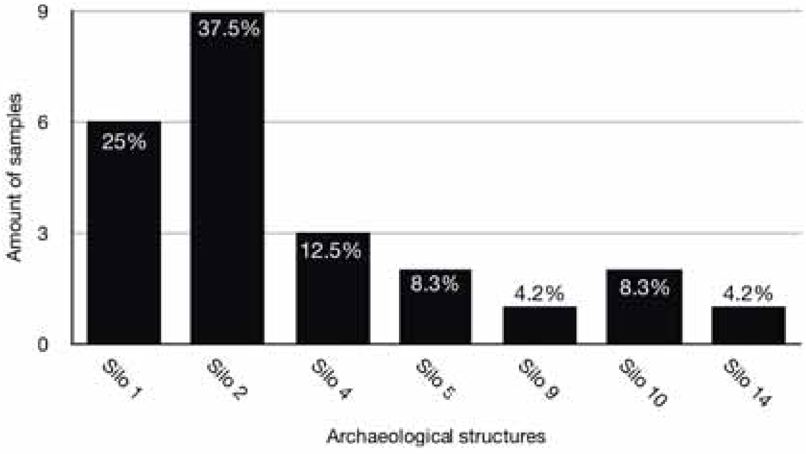


Figure 21: Amount of samples selected from each Early Neolithic structure. Percentages have been calculated from the total quantity of potsherds sampled.

The selected assemblage of 24 samples was composed of 25% rims (n=6) and 75% walls (n=18). Around 33% of the sherds had been fired under oxidised (n= 8) and 66% under mixed (n=16) atmospheres. Additionally, around 21% (n=5) of the samples were decorated. In two cases (CSP 47 and CSP 15), a series of linear incisions and, in another two cases (CSP 41 and CSP 49), a plastic application had been practiced in the rim offering a possible gripping feature. Samples CSP 20 and 15 also featured exterior plain appliqués in their walls and sample 47 presented the remains of what could have been a handle (Table 22).

The three samples originating from silos 9 and 10 (Table 22) are associated with the oldest dates in the site and placed right at the beginning of the second half of the VIth millennia BC (Gómez and Molist 2017). Alternatively, the remaining vases were located mainly in silos whose ^{14}C dates indicate the presence of debris from some point in the last three centuries of the VIth millennia BC. This chronological difference offers the opportunity to evaluate the presence of possible changes on the uses of pottery across time.

After removal of the interior surface, roughly 1g of pottery was drilled and lipids were extracted following the acidified methanol procedure and identified by GC-MS. In selected samples, compound specific isotopic values were obtained to further evaluate the origin of the residues. Work on these samples was performed in the British Museum (HTGC-MS) and the ICTA-UAB laboratories (GC-C-IRMS). Fatty acids in soils were analysed in two Postcardial structures, burial 19 and in hearth 17. This work was carried out in the University of Bradford.

Lab ID	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Possible interpretation
ABB1	39.5	1.6	-	-	-	Degraded animal fat
ABB2	45.0	0.9	-	-	-	Degraded animal fat
ABB3	50.7	1.1	-	-	-	Degraded animal fat
ABB4	7.4	1.0	-	-	-	Degraded animal fat
ABB5	10.7	1.2	-	-	-	Degraded animal fat
ABB6	105.6	0.5	-	-	-	Degraded animal fat
CSP10	245.8	0.8	-	-	-	Degraded animal fat
CSP11	14.4	1.5	-	-	-	Degraded animal fat
CSP12	15.8	1.1	-25.5	-24.9	0.7	Degraded non-ruminant adipose fat
CSP13	22.4	0.8	-25.3	-24.0	1.3	Degraded non-ruminant adipose fat
CSP7	36.0	1.5	-	-	-	Degraded animal fat
CSP8	91.6	0.3	-	-	-	Degraded animal fat
CSP9	20.1	0.8	-25.5	-26.0	-0.5	Degraded ruminant adipose fat
CSPA	9.7	1.0	-26.6	-27.5	-0.9	Degraded ruminant adipose fat
CSPB	12.1	0.8	-26.0	-25.0	1.1	Degraded non-ruminant adipose fat
CSPC	27.7	1.0	-27.7	-28.2	-0.5	Degraded ruminant adipose fat
CSPD	12.1	1.1	-	-	-	Degraded animal fat
CSPE	9.2	1.8	-	-	-	Degraded animal fat
CSPF	18.5	1.4	-25.4	-27.3	-1.9	Degraded ruminant adipose fat
CSPG	7.7	1.4	-	-	-	Degraded animal fat
CSPH	14.3	1.5	-	-	-	Degraded animal fat
CSPI	2.2	1.7	-	-	-	Non significant residue
CSPJ	16.7	1.0	-	-	-	Degraded animal fat
CSPK	3.1	1.6	-	-	-	Non significant residue
Burial 19	4.8	0.6	-	-	-	Soil lipids
Hearth 17	5.1	0.5	-	-	-	Soil lipids

Table 23: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in µg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio.

4.2.5 Results and discussion

Analysed DCM/MeOH extracts from Burial 19 and Hearth 17 in the Postcardial period only yielded minor amounts of organic compounds. In the case of Burial 19 only trace amounts of short alkanes were detected. In hearth 17, trace amounts of palmitic and stearic acid were considered to be lower than 1µg·g^{-1} and, therefore, incapable of generating the lipid signals detected inside the pottery matrices. To evaluate the potential presence of other bound lipids, an acidified methanol extraction was also practiced in both samples. In this case, higher amounts of lipids were detected. Nevertheless, the total lipid extracts were in both cases lower than 6µg·g^{-1}, thus supporting the 5µg·g^{-1} threshold stablished to reject residues which could have been significantly affected by soil lipid contamination. These results are coherent with previous analytical work indicating soil contamination could be minimal (Heron *et al.* 1991).

Analysis of recovered residues from pottery in Caserna de Sant Pau revealed significant amounts of organic molecules in 22 samples (91.6%) (Table 23). Overall, total lipid extracts (TLE) were exponentially distributed (median 16.25 $\mu g \cdot g^{-1}$) with a significant amount of samples containing between 0 and 25 $\mu g \cdot g^{-1}$ (n=16, 66%), fewer containing between 25 and 50 $\mu g \cdot g^{-1}$ (n=5, 20.8%) and only three samples with TLEs higher than 50 $\mu g \cdot g^{-1}$ (12.5%). It does not appear that either the firing conditions (Mann-Withney U=51.5 n=24 p=0.46), the sherd shape (Mann-Withney U=36 N=24, p=0.24) or the presence of decoration (Mann-Withney U=40 N=24 p=0.61) could be associated with statistically meaningful differences in the amount of lipids recovered. These results suggest fats were not differentially preserved along the vessel and that the vessel's firing technique did not affect the intensity of the lipid signal. Moreover, the presence of decorative or functional elements such as handles, appliqués or incisions did not seem to be associated with higher amounts of residues either. Alternatively, it remains unclear wether the presence of higher or lower amounts of organic molecules could be linked with the nature of the residue itself.

Although it seems that the TLE and the relative amount of palmitic to stearic acid (P/S ratio) could be correlated (see Figure 22) (Spearman's D= 3502 n=24 p=0.01), when sample CSP10 (not shown in Figure 22) is removed as an outlier and the two samples with non-significant amounts of lipids are not taken into account the correlation becomes most statistically significant (Spearman's D=2055 n=24 p=0.13). Furthermore, a linear regression on the available data generates a r^2 value around 0.3, thus suggesting much of the variability in the PS ratio is not affected by the quantity of lipids recovered in Caserna de Sant Pau.

Regarding the nature of the recovered lipids, all samples except for CSP15 contained detectable quantities of phthalate esters, a range of alkanes and volatile poliaromatic hydrocarbons which could originate from modern post-excavation contamination due to long term storage in plastic bags. The range of fatty acids present mainly spanned between C12:0 to C28:0 maximising at the C16:0 and C18:0 moieties. The presence of dicarboxylic fatty acids such as azelaic acid and, in some cases, keto acids is indicative of the post-depositional oxidation processes which affected unsaturated compounds. Biomarkers for the presence of marine products were not identified in any of the samples in Caserna de Sant Pau. Furthermore, the absence of wax esters either complete or possibly hydrolised (combination of very long chain alkanes and alcohols), plant specific sterols or odd over even quantities of alkanes prevent the clear

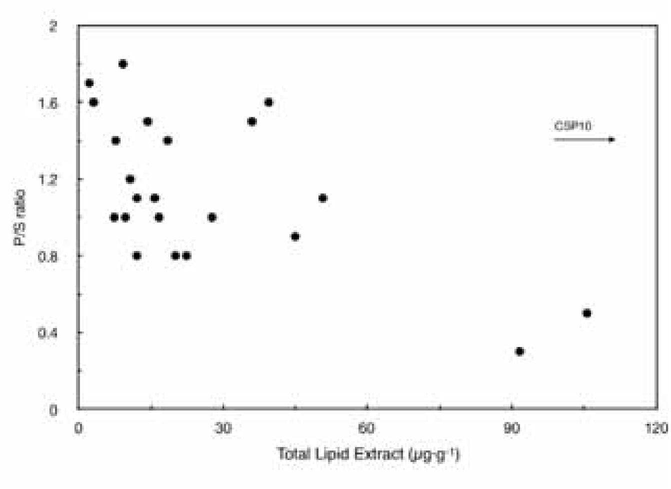


Figure 22: Possible correlation between P/S ratios and Total Lipid Extracts (TLE) in Caserna de Sant Pau. Sample CS10 plots otuside the chart.

identification of any major input of plant derived products. Phytanic acid, which is a degradation product from phytol but can also be found in the rumen of animals was detected only in one sample (CSP10) and although minor quantities of molecules related to the possible presence of degraded resins (methyl-dehydroabietic acid, 7-oxo dehydroabietic acid and retene) were identified in a significant amount of samples (n= 10, 41.6%) their origin remains unclear. Moreover, Palmitic to Stearic ratios are lower than 2 in all studied cases in Caserna de Sant Pau. Following criteria laid out by Copley *et al.* (2005e) and Dunne *et al.* (2016) amongst others, it is highly unlikely the present range of fatty acid could have originated from the complete hydrolysis of plant triacylglicerols. Alternatively, these profiles are not incompatible with hydrolised animal triglycerides, a possibility supported by the detection of trace amounts of cholesterol in samples CSP10 and CSP8.

Furthermore, although charred animal bones were found in the site, the absence of molecules resulting from pyrolithic reactions on lipids (mid chain ketones and cyclic fatty acids) prevents the identification of cooking practices in pottery involving high temperatures (>250ºC)(Hansel *et al.* 2004, Evershed *et al.* 2008a).

Samples containing sufficient amounts of palmitic and stearic acid, low signal to noise rations and an absence of negligible presence of UCMs were submitted to compound specific isotopic analyses.

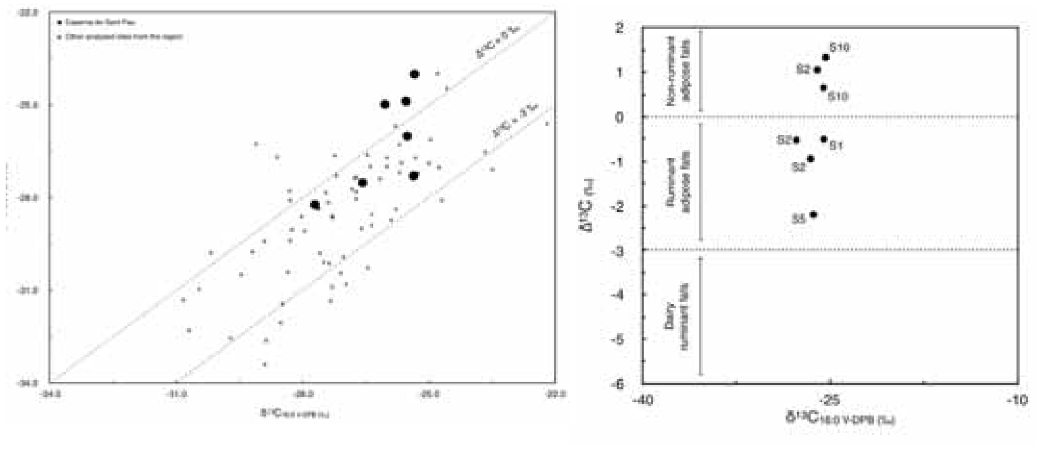


Figure 23: Results from compound specific isotopic studies from samples in Caserna de Sant Pau. a: xy plot with δ13C16:0‰ (x axis) and δ13C18:0‰ values (y axis). Black dots represent samples from this site and small crosses identify all isotopic values from the Early Neolithic available in the Iberian peninsula. b: plot presenting δ13C16:0‰ (x axis) and the Δ13C index (y axis), shown to minimise environmental variation and help better identify non-ruminant, ruminant and dairy fats.

Compound specific δ13C values for Caserna de Sant Pau reveal a significant amount of variability along the Δ13C index (Figure 23). In the cases of CSP13, CSP12 and CSPB, a significantly more isotopically enriched stearic than palmitic acid is not compatible with a significant input of animal fats from a ruminant origin. Marine oils, although sometimes similarly enriched, could also be ruled out because of the absence of specific biomarkers. Moreover, the absolute isotopic values are not coherent with significant inputs from plant oils or highly herbivore non-ruminant animals. Alternatively, modern authentic porcine adipose fats have been show to present similar values and, in absence of other possible explanations, it seems safe to assume that these samples could have contained at least significant amount of porcine adipose fats. In one case (CSP9) although the Δ13C index is coherent with the presence of ruminant adipose fats, this sample plots close to the porcine area and, therefore, minor inputs of non-ruminant fats should not be ruled out.

Other analysed samples (CSPC, CSPA, CSPF) present Δ13C values below 0, which suggests a significant input of ruminant fats should not be rejected. Given that dairy fat reference values do not agree with these samples it is unlikely any significant amounts of this product had been contained in the vessel. In consequence it could be assumed that ruminant fats would have originated from the adipose tissue.

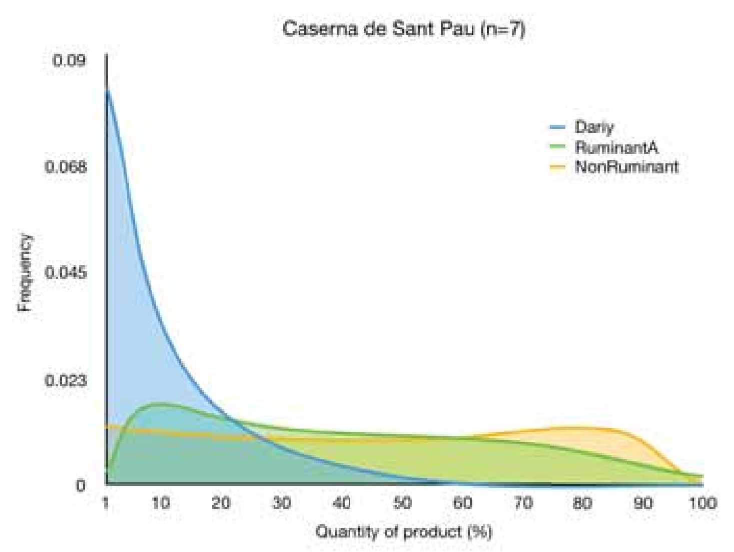


Figure 24: Modelled results averaging all samples isotopically studied in Caserna de Sant Pau.

To better assess the existence of possible mixtures and comprehensibly compare between samples across the VIth millennia, a bayesian mixing model (Fernandes *et al.* 2017) was applied to compound specific isotopic results. Given the absence specific biomarkers for plant or marine products, these were not taken into account. Alternatively, dairy, ruminant and non-ruminant reference adipose fats were included as end-members. The model works under the assumption that the palmitic and stearic isotopic values would result from fats accumulated across multiple vessel uses. Therefore, they could be assumed to represent the result of a weighted mean incorporating an ideal medium value for each type of fat, their standard deviation, and their amount in the mixture.

Model results over all samples analysed in Caserna de Sant Pau (Figure 24) suggest non-ruminant adipose fats could have been one of the main products processed and consumed in the site (median 47%) while the presence of significant quantities of ruminant adipose fats should not be rejected (median 37%). The presence of dairy products would only be coherent with the model results if quantities were minimal (median 9%). Nevertheless, this situation does not seem to hold when samples from different time periods are analysed separately.

Estimates from the two samples in silo 10 both are coherent with the presence of mainly non-ruminant adipose fats (median 74%) (Figure 25a). The low probabilities attributed to ruminant adipose and dairy fats (18% and 5% respectively)

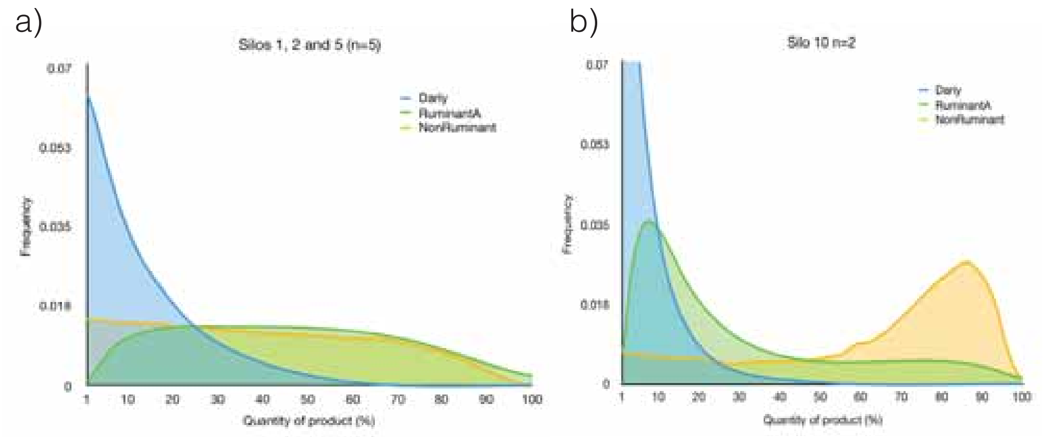


Figure 25: Modelled results averaging all samples isotopically studied from a) Silos 1,2 and 5, dated around 5300-500 cal BC. and b) silo 10, dated between 5534-5380 cal BC. See table x.

suggests it is not possible to assume these values could have originated from extensive mixtures of different products. Alternatively, non-ruminant adipose fats could have been almost exclusively used in these cases, which hints at the possibility that these were specialised vessels. Data from the remaining silos still presents significant probabilities of containing a certain amount of non-ruminant fats (median 38%) (Figure 25b), but these are significantly lower and closer to the expected quantities of ruminant adipose fats (median 44%). The one constant across time in Caserna de Sant Pau would be the minimal presence or absence of dairy products. Mann-Withney statistical tests evaluating whether the detected differences are statistically significant suggest that the null hypothesis than the distributions are similar can be rejected in all three cases (non-ruminant: U=4025, p=0.017, ruminant: U=3557.5, p=0.0004, dairy: U=3735, p=0.00019).

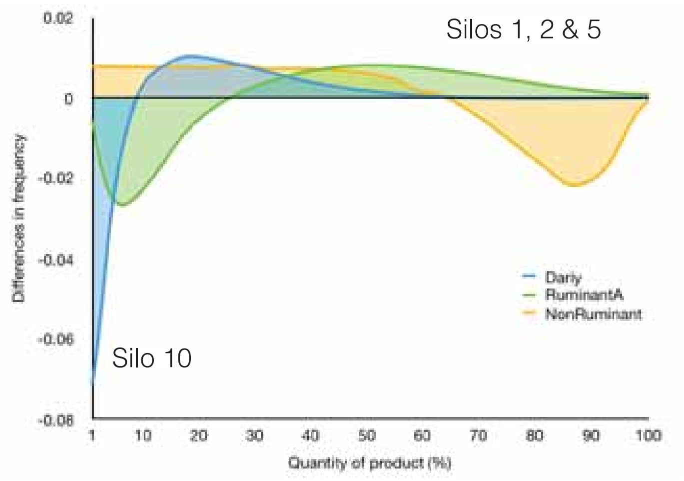


Figure 26: Differences in the statistical distributions between silo 10 and silos 1, 2 and 5.

Going beyond the study of the contained products, although the specific dimensions of ceramic vases might be difficult to obtain, the width and rim diameter of some of the analysed rims from silos 1 and 2 suggests these could have been rather big vessels (CSP47 and CSP15). The presence of significant amounts of possible degraded animal fats in both cases could possibly be related to the storage of food using fat as a protection. Nevertheless, experiments reproducing the incorporation of organic residues in the clay matrix have detected that heat facilitates the mobilisation of animal fats and, thus, increases their quantity in the vessel (Charters *et al.* 1997). Therefore, although the absence of molecules with a pyrolithic origin makes this last option less clear, the possibility that food was prepared in significant quantities at once in these vessels should not be fully rejected.

Organic residues from Caserna de Sant Pau originate from two clusters of non contemporaneous silos. Silos 9 and 10 present the oldest dates for the existence of Neolithic societies in the Barcelona plain. Given that only three samples could be analysed any results must be interpreted with caution. Yet, following Colominas *et al.* (2008), silo 9 seemed to contain slightly more pig remains that the remaining silos with the presence of animal bones (1 and 2). A humerus distal epiphysis belonging to a pig was also identified in silo 2, but no porcine remains were detected in silo 1. Looking at the residues, one of the three vessels whose isotopic values could be associated with pig adipose fats was indeed from silo 2 and the other two were from silo 10. Although no bone remains were found on these last feature, the fact that it is chronologically closest to silo 9 and half of the animal bones from that silo (N=3) belonged to pigs could be seen as a tendency linking the existence of Sus sp. bones with porcine residues. Moreover, the isotopic values from the single vessel analysed from silo 1, which had no identified pig remains, does plot in the ruminant area. This possible relationship is extremely weak given the small amount of residues and bones available and will need to be confirmed in future studies. Nevertheless, the data acquired so far does not reject that Sus sp. were consumed right since the beginning of the Neolithic in Caserna de Sant Pau, possibly in function specific vessels.

4.2.6 Conclusion

In conclusion, organic residue analysis on pottery samples from the Early Neolithic silos in Caserna de Sant Pau revealed vessels could had been used to process animal fats, possibly to prepare them for human consumption. The absence of sufficient data in these cases regarding other products such as dairy, marine or plant fats suggest these were either not well preserved or not used or used in small quantities. Further isotopic studies have been able to distinguish the existence of vessels mainly used for the processing of ruminant or non-ruminant (most possibly porcine fats) products. Although the amount of samples is still small, it seems that these could be in agreement with the animal bone assemblage recovered from the same archaeological contexts.

4.3 Carrer Reina Amàlia

4.3.1 Introduction

Carrer Reina Amàlia 31-33 (CRA) is in the Raval quarter in Barcelona, around 5 meters above the sea level and less than 1km from the sea. Its exceptionality comes from the fact that it is the oldest known domestic building on the Barcelona plain. This site is a clear example of the consolidation of agro-pastoral societies and one of the few fully excavated archaeological contexts of this type. Therefore, it offers a clear opportunity to further explore the development of the first Neolithic and obtain new data on the uses of the Epicardial vessels.

CRA is closely surrounded by a set of nearby archaeological sites such as C/Nou de la Rambla, Caserna de Sant Pau del Camp or C/Beates 2, which presented chronologies that suggest that the Barcelona Plain was continuously occupied since as early as 5615-5482 cal BC (González *et al.* 2017) and through the early, middle and late Neolithic. In fact, this whole cluster should be regarded as a

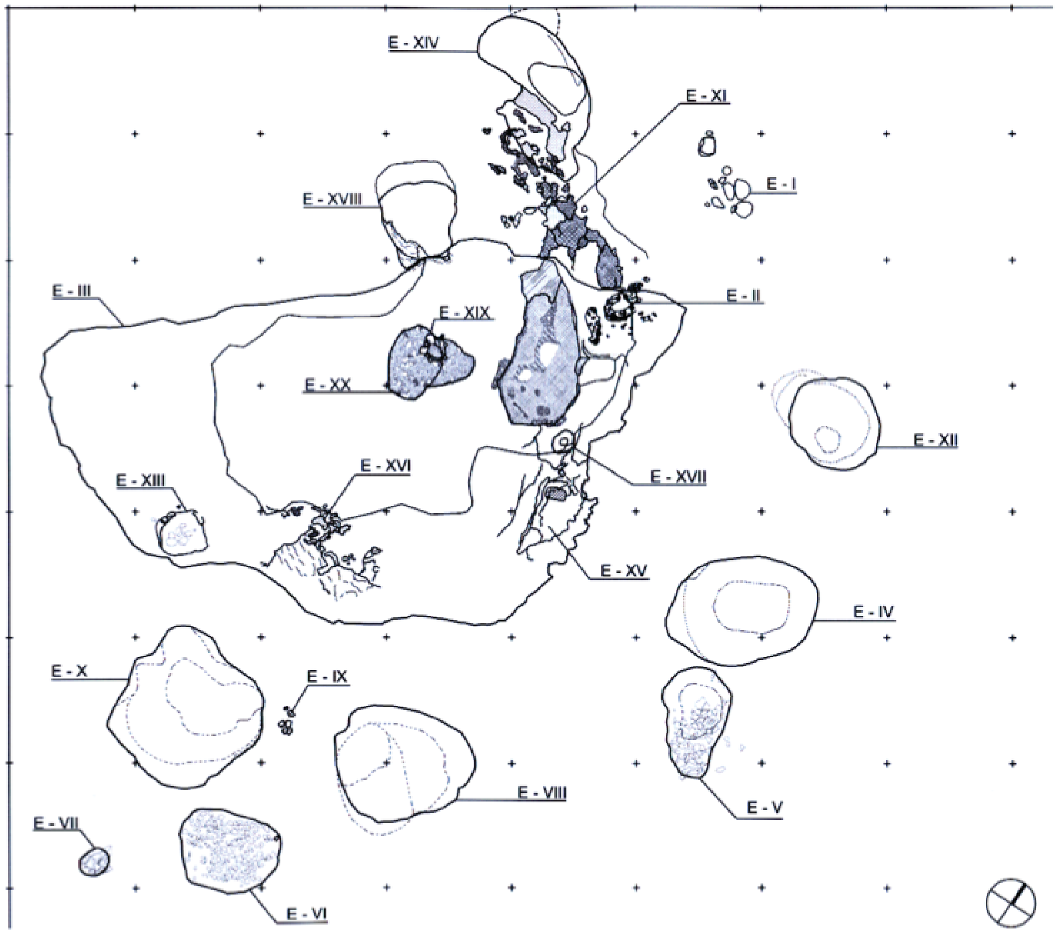


Figure 27: Plan of the Early Neolithic structures in Carrer Reina Amàlia.

single site instead of a set of individual excavations named after the streets of Barcelona today. When perceived this way, CRA presents many of the most important features of an important Neolithic settlement in the centre of the Catalan coast line.

4.3.2 Archaeological features relevant in this study

The site concealed a set of negative structures including three burials (E-XIV, E-XV and E-XVI), two post holes (E-XVII), five possible silos (E-IV, E-VIII, E-XII and E-XVIII) and one big shallow cut (E-III) that, because of the presence of a hearth inside (XX), has been interpreted as a domestic area. The latter has provided the vast majority of the findings and the stratigraphic sequence of the site. As depicted in Figure 27, the circular distribution of the structures suggests that other smaller pits were somehow related to E-III. They possibly acted as auxiliary spaces for storing products and supporting domestic life. In fact, the three burials have been also spatially related to the domestic space. The first two are located inside E-III and both contained child inhumations (1.5 and 4 to 5 years old respectively). Outside but in proximity, the third burial presented a double chamber structure with Chassey-type pottery. As confirmed by a ^{14}C date, the presence of this artefact suggests a more recent chronology.

In terms of stratigraphy, the site rests on top of clay deposits of brown and reddish colours. The presence of carbonates, sands, quartz and slate gravels in this matrix suggests that the sediments date from the Upper Pleistocene (González and Harzbecher 2008). These were formed by alluviation of materials coming from the Collserola chain, which limits the Barcelona plain. This same phenomenon generated important post-depositional effects in the site. Moreover, the analysis of the stratigraphy, the archaeological structures and the recovered artefacts allowed the excavators to structure the site in four main phases from the oldest to the most recent.

Phase I marks the construction and use of E-III, characterised by the stratigraphic unit (SU) 95 (González and Harzbecher 2008, González *et al.* 2011, Harzbecher and González 2016). The presence of a hearth made of heat fractured stones, thermally altered sediments and ashes, suggests the space had a domestic function. Significant numbers of cremated animal bones have also been found. Pottery fragments decorated with orthogonal appliqués indicate that this layer dates from the Epicardial period (5000-4500 cal BC (Martín *et al.* 2010). A ^{14}C date from a pig bone in the hearth deposits has provided a bayesian modelled date between 4667 and 4539 years cal BC and another pig bone from SU95 dated from 4658 to 4537 cal BC (González *et al.* 2017), which corresponds to the same time span that the pottery decoration suggests. This phase is largely covered by an irregular calcium carbonate layer, which is mixed with the lowest levels of phase II.

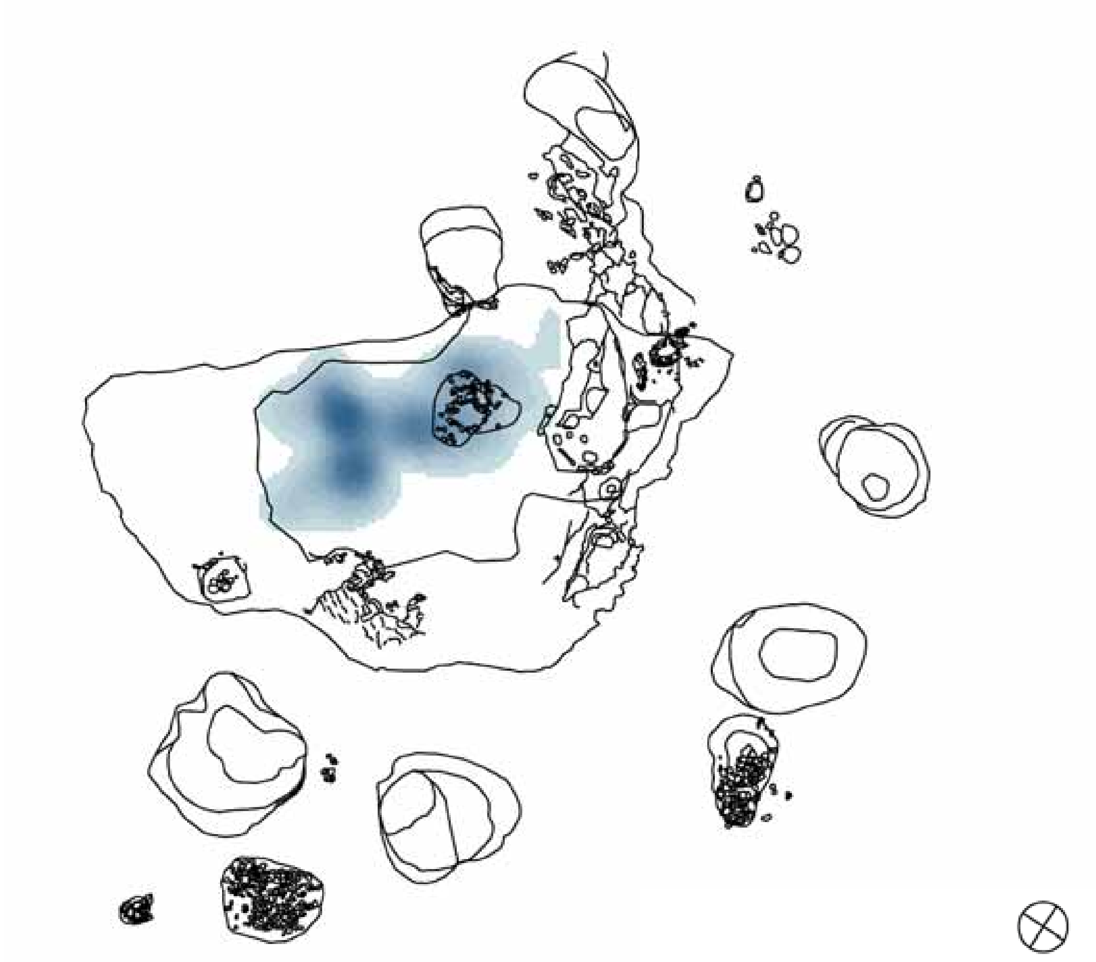


Figure 28: Interpolation showing the distribution and concentration of archaeological finds within phase I.

The kernel density map in Figure 28 shows the quantity and distribution of archaeological finds associated to this phase within the site. It would seem that the archaeological material would be concentrated around two focus, the first one by the hearth and the second one, to the South-West.

Phase II comprises many strata such as SU81 or its main, SU59 (González and Harzbecher 2008). A ^{14}C date on a bovine rib locates this layer between 4576 and 4500 cal BC (González *et al.* 2017). This phase overlays two child burials

(E-XV, Burial I, and E-XVI, Burial II) and a posthole (E-XVII), which could have been in use by this phase but were cut into the Upper Pleistocene deposits. Human bone from Burial I was ^{14}C dated producing a bayesian modelled calendar date from 4590 to 4511 years cal BC (González and Harzbecher 2008).

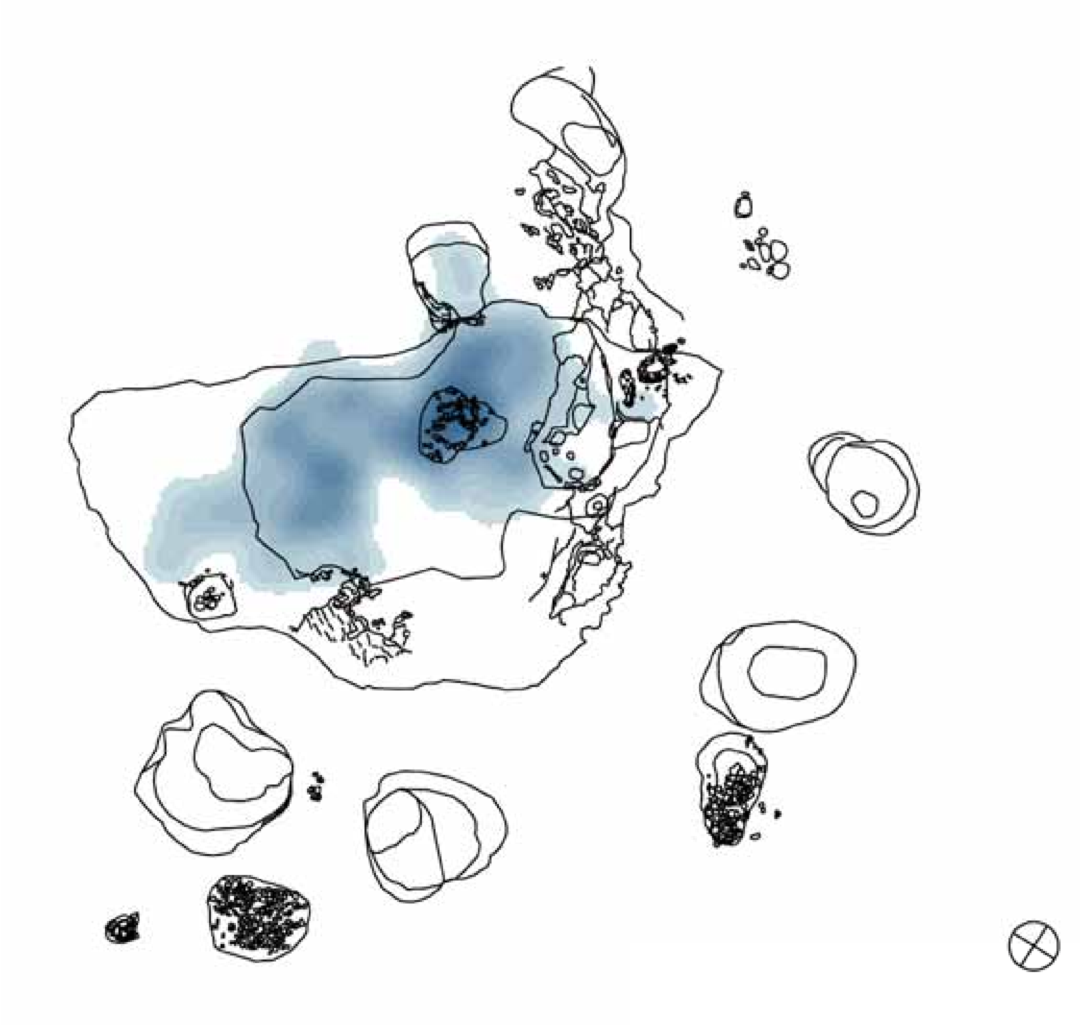


Figure 29: Interpolation showing the distribution and concentration of archaeological finds within phase II

Within this phase, an assemblage of 1180 pottery sherds was excavated. The decoration was composed by vertical, horizontal and orthogonal appliqués, which match the style of Phase I and suggest an Epicardial date. Seashells and the lithic industry, including jasper, were indicative of the economic activities that this group performed in exploiting their environment. The spatial distribution of the archaeological remains associated with this phase directly overlay the ones from phase I and cover a larger area. The presence of two clusters of remains in the same disposition is still valid for this phase.

Phase III is the last one in E-III. It is mainly composed by SU46 (González and Harzbecher 2008) and presents a change in pottery decoration that also indicates a change in chronology. A small portion of the 1500 sherds from this phase possessed a set of features such as tubular handles and carinations (González and Harzbecher 2008). These markers point to the preceding period, the Postcardial, an interpretation coherent with a bovid rib bayesian modelled ^{14}C date between 4569 to 4482 cal BC. During this time, the Neolithic society was transforming in a way which could have caused the abandonment of the site. The kernel density distribution of archaeological remains in the site shows that this phase would have covered an even larger area but with a significant change in the disposition of the objects compared with the two previous phases. As depicted in Figure 30, the distribution would be now a continuous spread from approximately the North-East to the South-West.

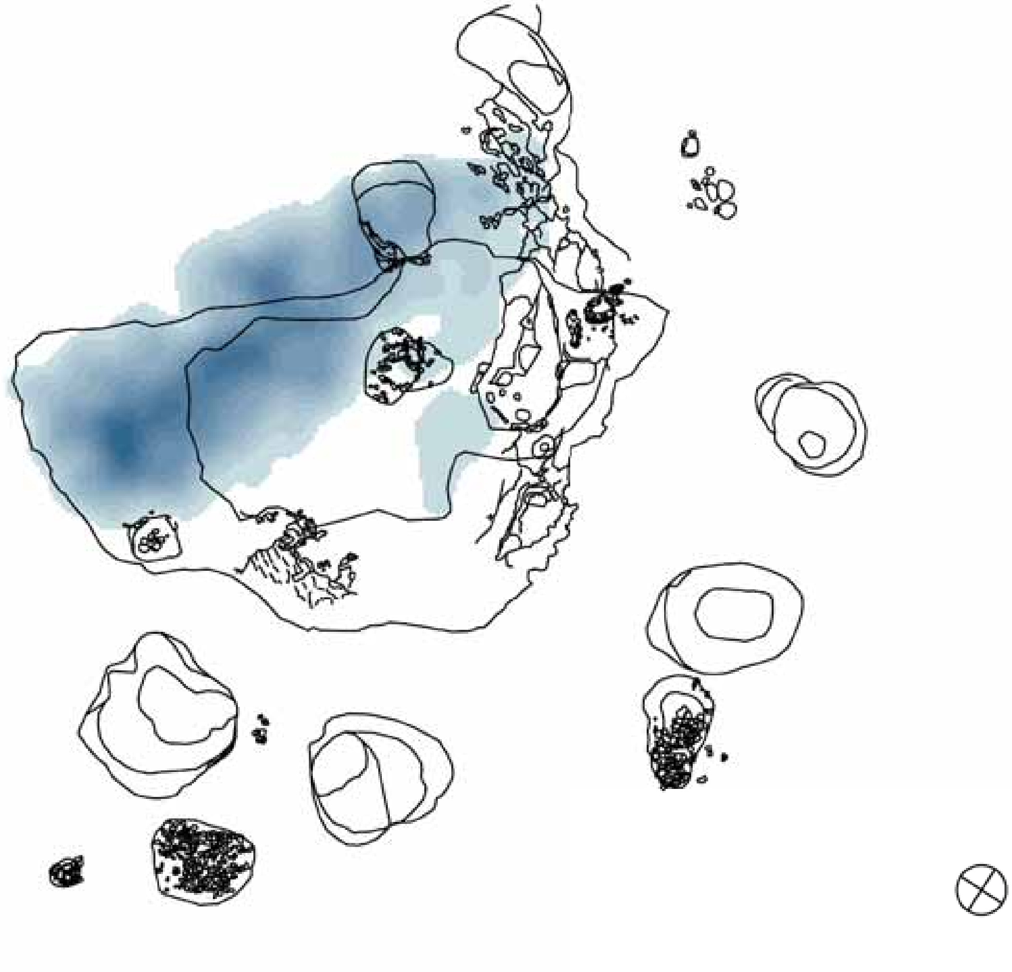


Figure 30: Interpolation showing the distribution and concentration of archaeological finds within phase III

Phase IV is the most recent phase in the site and indicates the covering of the archaeological structures by alluviation. Stratigraphic units such as SU15 sealed E-III (González and Harzbecher 2008) and E-IV. In fact, a ^{14}C date from a human bone (Burial III in E-IV) provided a calendar date from 4530 to 4360 years cal BC at 2σ. Such a result would securely place these layers beyond the Epicardial period and would suggest that the main occupation of the site was, indeed, between 4700 and 4500 years cal BC. The distribution of the archaeological remains in Figure 31, still shows a concentration overlaying the previous materials from phase III but shows the spread of materials to be located across all the structures in the site.

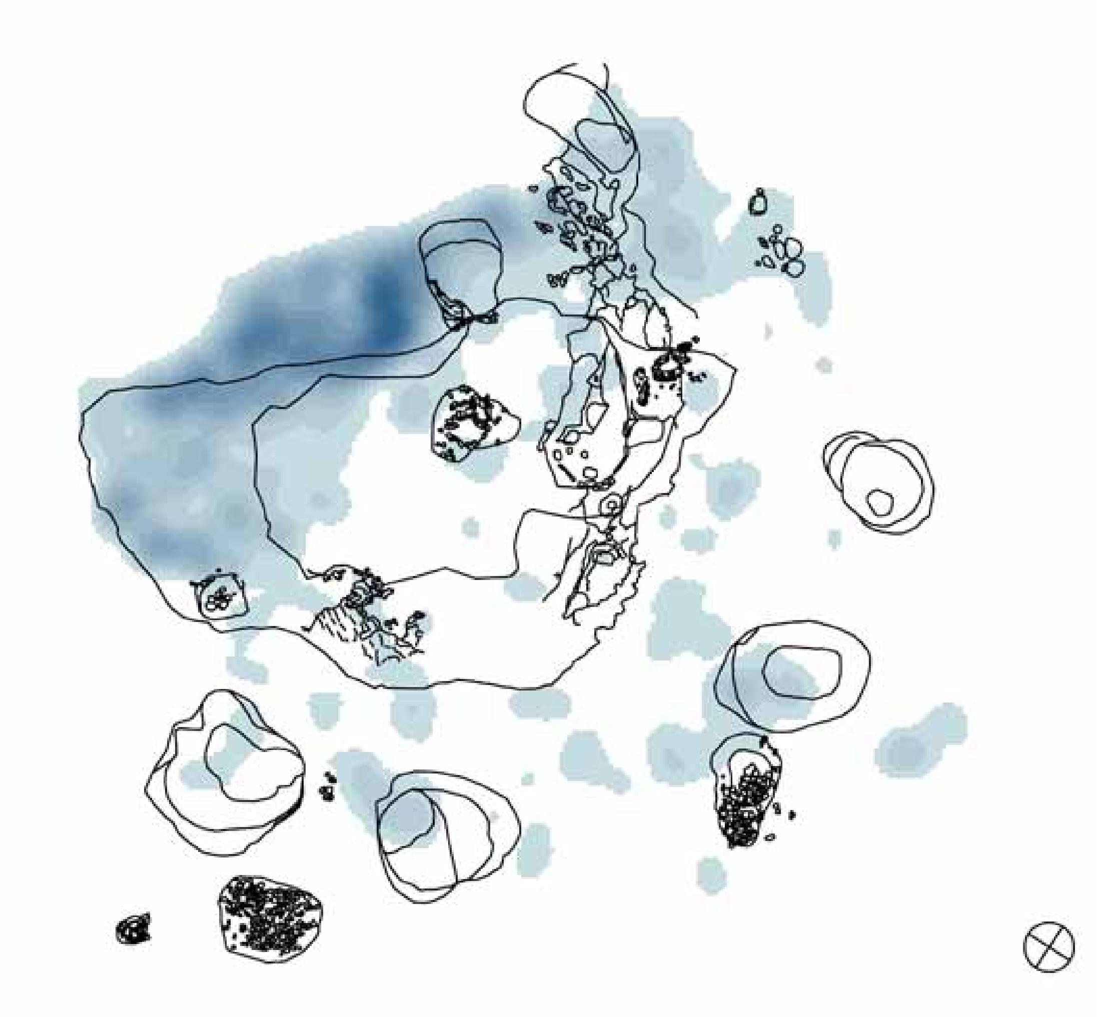


Figure 31: Interpolation showing the distribution and concentration of archaeological finds within phase IV.

A Bayesian model using the eight available dates in the hut's stratigraphy suggests the structure's domestic use could have lasted less than half a century. Additionally, the transformation of the space to a burial place and the refilling processes would have lasted no more than 150 years. These results are conditioned by the fact that the date from the first abandonment (SU95) is highly similar from the hearth date. The possibility that further ^{14}C analyses on charred seeds or other bones from the hearth provide older dates should not be ruled out. In fact, a ^{14}C charcoal date from feature XII, a hearth outside the hut, yielded a date from 4827 to 4692 cal BC at 2σ. Although the possibility that an old wood effect is skewing this result is real, this also implies further work could fill the current chronological gap between the latest dates at Caserna de Sant Pau and the youngest dates at Carrer Reina Amàlia.

4.3.3 Archaeological materials

4.3.3.1 Pottery remains

In terms of finds, the main bulk is situated between 5.29 and 3.80 meters above sea level. Regarding pottery, 553 of the 6246 recovered sherds were decorated. Their shapes included middle to big sized containers and globular shapes (Gómez 2016) The main decorative technique was the use of appliqués (72.3%), although some other minor techniques such as impressions (0.7%) and incisions (1.2%) were also present. The latter slightly increased in frequency over time. Carinations and tubular handles were exclusive of phase III, but many other characteristics appear to be widespread all along the stratigraphy of the E-III structure. Rims were analysed using the Dedet-Py categories (Dedet and Py 1975), and suggested that no major shape differences existed between these three phases. Only minor variations should be noted between Phase I and II, with a slight increase in the frequency of closed rims.

In terms of the production process, the colouration of the pot is an indicator of the firing environment. Using this principle, 30% of all recovered sherds from E-III were fired under reductive conditions, 64% presented a mixed colouration that suggested fluctuation in the availability of oxygen, and, finally, only 6% of the fragments had experienced oxidation. This distribution clearly indicates that the firing technology available did not include the control the amount of oxygen entering the kiln. Colouration should have a minor impact in the functionality of the vessels, but since different cooking atmospheres triggered different chemical reactions, the range of minerals present in one or the other might be significantly different. Further details on the pottery assemblage can be found in appendix 3.

In terms of post-depositional processes, the degree of fragmentation did not seem to be as high as in other archaeological sites. Although no complete vessels were recovered, 20% of the sherds preserving parts of the vessel shape and pieces that weighed more that 100g were abundant. As previously stated, it was common in this site to find calcium carbonate in the form of a crust (González *et al.* 2011). It is not unusual to find these encrustations as coatings on the interior and exterior surfaces of approximately 15% of the pots. These crusts are also an indication of chemical processes that were active when the sherds were buried. Therefore, these reactions should be considered when archaeometric analysis are performed.

4.3.3.2 Lithic remains

Regarding lithic tools, up to 2056 pieces were recovered across the site. The majority of them had been prepared on flint from two possible sources although jasper, chert and quartz had also been used (González and Harzbecher 2008). The presence of cores, flakes, blades and retouched pieces including a series of microliths suggests some but not all lithic tools had been knapped in the site (González and Harzbecher 2008). Macrolithic remains included grinding instruments and polished tools (González and Harzbecher 2008, Bofill 2016).

4.3.3.3 Animal and human remains

Animal bones found in all features were analysed by Vanessa Navarrete and Roger Molinas. Up to 4129 animal bone remains (Molinas 2009) which allows a statistically relevant interpretation of the herding practices, were found across all features. The site consumed mainly domesticated animals. Although *Bos taurus* (49.05%) appeared to be the most abundant species, followed by ovicaprines, a significant amount of pig remains was also present (28.81%). The MNI values implied that ruminant animals could have been the main source of protein intake. In terms of herd management strategies, the immature/mature rate suggests that animals could have been raised for meat consumption. Cattle were usually slaughtered between 2 to 3 years old; pigs between 18 and 36 months of age; sheeps between 1 and 3 years, and goats between 2 to 3 years. According to the researchers, this seems to be a polyvalent composition which could provide meat and also secondary products such as milk or field labour (Saña and Navarrete, 2016).

Recent work by Navarrete *et al.* (2017) studied the δ^{13}C and δ^{15}N bulk collagen values in suid bones from Carrer Reina Amàlia. When results were compared to herbivores, similar δ^{13}C values validated that suids could have consumed C3 plants or C3 fed animal products. Regarding δ^{15}N values, pigs were on average 2.5% enriched compared to on-site herbivores. This suggested it should not be ruled out that a relatively higher proportion of animal sources contributed to their dietary proteins. The researchers indicated that this feeding practice could be associated with home-based management systems where pigs would have been enclosed or relatively free to forage only within the settlement, thus increasing the possibilities of ingesting animal foodstuffs. These studies treated pig bones in Caserna de Sant Pau and Carrer Reina Amàlia as originating from the same site due to the close proximity of the excavations within the city of Barcelona.

Marine remains included a set of 534 seashells (Nadal *et al.* 2015). The vast majority of these were either *Glycimeris sp.* (NMI = 40%) or *Patella sp.* (NMI = 38.3%). The remaining 21.7% was composed of a series of species with only minor amounts of remains each (ex: Stramonita haemastoma or Acanthocardia tuberculata)(see Nadal *et al.* 2015 for a full report). Although the majority of the Glycimeris and other species were highly weathered and, therefore, not associable of human consumption, researchers did not rule out the possibility that the remains of *Patellae* could have been part of the human diet. Moreover, a small amount of fish remains including bones were detected in the site and some macrolithic tools have been interpreted as possible fishing-net weights (Gómez *et al.* 2014b, Nadal 2016). Apart from this possible use as a food source, the malachological assemblage has also been interpreted as the remains of the raw materials resulting from the production of personal attire.

Finally, the study of the human remains found in the site suggested that the buried children could have possible been feeding with solids since an early age due to the presence of dental calculus. It is therefore possible that these individuals would have been weaned really early, but the absence of dentine hypoplasia and δ^{15}N collagen values prevents any further interpretation of the possible reasons for such phenomenon (Malgosa 2016). Alternatively, the δ^{13}C value provided with the ^{14}C date of one of the individuals does not present values coherent with a significant consumption of marine products (-19.1‰). Although indicative, this result needs to be further evaluated through a complete isotopic palaeodietetic study. Overall, the study of the human remains from the Cardial, Epicardial and Postcardial phases in Caserna de Sant Pau, Carrer Reina Amàlia and Plaça Vila de Madrid suggests that the presence of marine products in the diet should not be completely ruled out and the low presence of dental pathologies could suggest the presence of a diet still not fully dominated by carbohydrates (Malgosa 2016).

4.3.3.4 Plant remains

Studies by Ferran Antolín and Anna Rodríguez recovered 589 seeds and fruit remains from features across the site (E-I, E-II, E-III, E-XI, E-XII, E-XVI, E-XVIII and E-XX). In the Epicardial phases, remains were mainly cereals such as naked barely, naked wheat and emmer. The total 142 remains from this phase also included some ruderal plants and one fruit, *Pistacia lentiscus*. A significant 447 remains were found in the features associated with the second half of the fifth millennia BC. These included mainly cereals such as *Triticum aestivum/durum* and *Hordeum vulgare*. Overall, botanic remains from the site suggest societies at the end of the Early Neolithic in the Barcelona practiced a floodplain agriculture involving free-threshing cereals. For more detail, see Antolin's full report (Antolín 2013).

Research led by Riera *et al.* (2016) and Mensua and Piqué (2008) has also provided evidence of the vegetal environment in the Barcelona plain. The studies suggest woods were mainly composed of a mixture of riparian and mixed oak trees. Data from Caserna de Sant Pau provides a nearby context suggesting the existence of Mediterranean trees with other supramediterranean taxa. In summary, *Quercus sp.* would have been the best represented species as riverside types were absent from the studied record. Data from cores practiced across the plain suggests that the Barcelona seashore would have witnessed a raise in the sea level at the onset of the Neolithic (Julià and Riera 2012) which would have conditioned the development of humid climate conditions with the presence of freshwater marshes. This paleoenvironmental setting could have changed around 4700-4500 to a more arid situation. This transition would be starting at the moment when Carrer Reina Amàlia was being occupied.

As presented, CRA revealed a complete dataset able to significantly contribute to the knowledge of Neolithic societies. The presence of a vast bone assemblage with lithic industry, seashells and plenty of pottery will help shed new light on the Epicardial period. When attempting to understand the processes of change in these societies, the study of those elements which present variation must be the starting point of enquiry. In the case of the Cardial-Epicardial period, one the artefact characteristics that seems to have changed the most is pottery decoration (Ten 1979). To better understand possible changes in pottery production, this artefact has to be also seen as an active agent. It is possible that, since the characteristics of pottery changed, its purpose could have changed as well. Therefore, a set of questions should be considered. What did these pots contain? Were differences in shape and/or decoration related to the contents of the vessels? If pottery changed, did its use also change? If these questions are to be further explored, new data should be acquired about the actual contents of the vessels. One of the main and more successful techniques for accessing this information is organic residue analysis.

4.3.4 Sampling and analytical techniques

To this end, 50 rims, 1 base and 1 wall, (19 sherds from phase I (layer 95) and 33 sherds from phase II (layer 59), were selected for analysis. Although the presence of extensive calcium carbonate crusts prevented a full study, in both contexts, surfaces were mainly smoothed and only in some cases some polishing could be observed. The firing technology was coherent with the expected in the period and temper included abundant quartz and mica minerals amongst others. In terms of the physical characteristics of the studied sherds, the thickness, diameter of the rim and the aperture were used to differentiate between different shapes. Results are presented as percentages in Table 24. For more detail on each sample see appendix 3.

	59 (%)	95 (%)
Thickness		
6-9	12.1	15.7
9-12	48.5	52.6
12-16	39.4	31.5
Diameter		
20-30	20	50
31-40	13.3	50
41-50	66.7	0
Aperture		
Open	35.5	6.6
Straight	48.4	33.3
Closed	16.1	60

	59 (%)	95 (%)
Decoration		
Yes	42.4	57.8
No	57.6	42.1
Firing		
Reductive	54.5	36.8
Mixed	39.4	52.6
Oxidative	6.1	10.5
Calcaric crust		
Yes	63.6	36.8
No	36.4	63.2

Table 24: Characteristics of the analysed vessels from layers 59 (Phase I) and 95 (Phase II) in Carrer Reina Amàlia.

It was possible to estimate vessel shape in 14 cases (26.9%) of the overall 52 studied sherds. Across both stratigraphic units, hemispheric vases were 42.8% of the assemblage, followed by subspheric (28.5%), necked (14.2%) and cylindrical (14.2%) vessels. The dimensions are one of the important physical characteristics in the functionality of vessels. Although the fragmentation of the samples made the direct calculation of volumetric capacity impossible, other variables were explored to approximate dimensions. Figure 32 presents the relation between rim diameter and body thickness. Following Henrickson and McDonald (1983) it has been proposed that cooking pots will have a height between 1 and 0.33 times the rim diameter. Furthermore, in samples from the same region and similar chronology (Bosch *et al.* 2011) vessel height is usually between 1 and 0.4 times the rim diameter. Using this data, an approximation to vessel capacity can be attempted using the formula for the volume of the cone.

$$V = \pi r^2 \frac{h}{3} \qquad \mathbf{9}$$

Therefore, in samples from context 59, two thirds of the studied assemblage would possibly have been able to contain at least 10l of a liquid. Conversely, the two vessels in unit 95 whose diameter is available would have been only able to contain less than 10l. These values could help understand the potential use of each vessel but they should not be considered accurate and therefore no statistical analysis have been performed with them. Finally, using the separation between two groups that can be appreciated in Figure 32, 30cm of rim diameter have been considered the threshold between smaller and larger vessels in this assemblage.

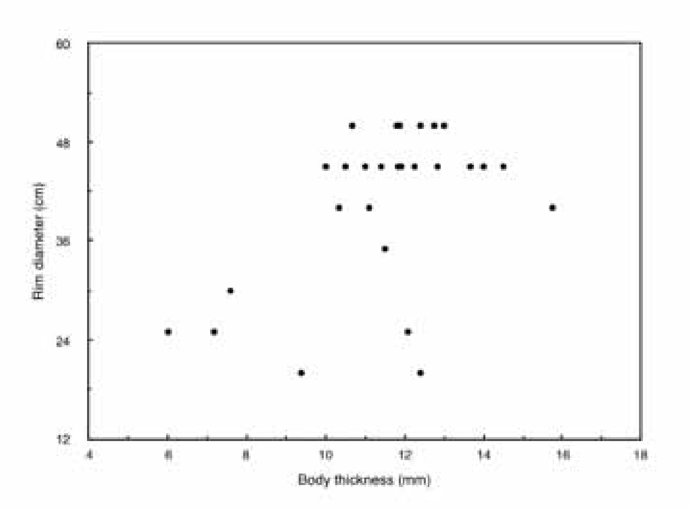


Figure 32: Plot comparing the rim diameter and body tickness in pottery sherds from stratigraphic unit 59 in phase II

Regarding decorations, appliqués were present in 48% of the samples and the remaining sherds presented either no decoration (42.3%) or other functional plastic implements such as handles (9.6%). These tended to be in a horizontal disposition although some cases show vertical (95/9092) and curved (59/6100, 95/9359) orientations. In the analysed sherds, they tended to be between 10 and 60cm wide and usually placed between 0.5 and 3cm below the rim, which does not exclude the presence of further appliqués lower in the vessel wall (59/6522, 59/7656). The placement of appliqués following an "T" shape is one of the characteristic traits of the epicardial assemblages (Manen 2002). Furthermore, five samples incorporated handles of varied types in the exterior surface (95/9565, 95/9189, 95/9630, 95/9680).

All samples were analysed in the University of Bradford via GC-MS after an acidified methanol extraction was practiced on around 1g of pulverised pottery. Compound specific isotopic values were obtained from instruments in two laboratories. Data on stratigraphic unit 59 was studied in the University of Liverpool and data on stratigraphic unit 95 was obtained from the GC-C-IRMS instrument in the ICTA laboratories in Barcelona.

4.3.5 Results and discussion

The TLE from the whole assemblage are presented in Table 25 and Table 26. Samples originating from phase I presented a wide range of lipid concentrations. All of the studied sherds contained more than 5 $\mu g \cdot g^{-1}$ and 10 $\mu g \cdot g^{-1}$ and 63% (n=12) had more than 100 $\mu g \cdot g^{-1}$. In phase II, while 57.6% of the sherds yielded quantities of fatty acids above 100 $\mu g \cdot g^{-1}$, 87.9% were above 5 $\mu g \cdot g^{-1}$, and 78.8% were above 10 $\mu g \cdot g^{-1}$. These two thresholds have been used in the literature (Copley *et al.* 2005a, Craig *et al.* 2005a, Craig *et al.* 2011, Dudd *et al.* 1999, Evershed *et al.* 2008b) to mark the presence of significant quantities of lipids that cannot be explained from soil and post-excavation contamination alone. Nevertheless, identification of the fats' origins could not be done in all samples due to the degree of degradation of the residue. Table 25 and 26 present the detected biomarkers. The absence of triacylglycerols and the presence of diacylglycerols and monoacylglycerols in DCM/MeOH extracts suggests that the fats were highly degraded. Moreover, the molecular profile of the residues indicates that the effects of both hydrolysis and oxidation reactions were extensive. A range of keto acids from C18:0 and dicarboxylic acids were detected. Table x in the appendix presents the range of oxidation products that have been found with

Lab id	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	APAA	CFAM	Phytanic acid
5561	1.55	0.8	-	-	-	0	0	0
8439	2.64	0.9	-	-	-	0	0	0
4892	3.8	0.6	-	-	-	0	0	0
8263	3.95	0.4	-	-	-	0	0	0
11960	6.72	0.8	-	-	-	0	0	0
7656	8.91	0.8	-	-	-	0	0	0
7718	9.52	1.4	-	-	-	0	0	0
8764	10.86	0.5	-	-	-	0	0	0
6162	16.69	1.2	-	-	-	0	0	1
8392	19.97	1.8	-	-	-	0	0	0
6522	20.62	1.2	-	-	-	0	0	0
8842	34.64	1.4	-	-	-	0	0	0
8009	40.96	1.6	-	-	-	0	0	0
9065	47.6	1.1	-	-	-	0	0	1
7778	114.88	1.2	-	-	-	0	0	1
8726	214.16	1.5	-25.7	-27.2	-1.6	1	1	1
5823	256.53	1.8	-	-	-	0	0	0
6231	319.44	1.4	-27.3	-30.9	-3.8	0	0	1
8721	333.73	1.2	-	-	-	0	0	0
9064	357.43	1.2	-	-	-	0	0	0
6681	373.57	1.9	-27.3	-28.6	-1.4	0	0	1
6725	387.68	1.4	-	-	-	0	0	1
5531	428.33	1.3	-28.3	-29.4	-1.2	0	1	1
5132	432.33	1.1	-27.6	-29.8	-2.3	0	0	1
8413	439.32	1.6	-	-	-	0	0	0
6100	456.96	1.6	-	-	-	0	0	1
9464	644.93	1.3	-	-	-	0	0	0
8070	742.3	1.0	-	-	-	0	0	1
6405	769.5	0.9	-	-	-	1	1	0
6695	778.74	1.3	-	-	-	1	1	1
8737	1028.11	1.4	-27.5	-30.1	-2.7	0	0	1
8234	1095.89	1.0	-	-	-	0	0	0
8995	1226.41	1.1	-	-	-	0	0	1

Table 25: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses in layer 59 (phase II). TLE: total lipid extract is expressed in µg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio. APAA: ω-(o-alkylphenyl) alkanoic acids. CFAM: other cyclic fatty acid monomers

its retention times In terms of the identification of the origins of the residues, several indicators have been taken into account. Firstly, the C16:0 and C18:0 peaks usually represent between 60% and 80% of the total amount of lipids recovered. The ratio of palmitic to stearic acid relative concentrations is in all cases between 0.5 and 2 and there seems to be no correlation between these values and the total lipid extracts (Spearman's D=23308, n=52, p=0.99). As it has been previously reported (Copley *et al.* 2003), these high quantities would not be against the presence of animal fat in the vessel. Nevertheless, both stearic and palmitic acid are the most common fatty acids in nature. Although they are

Lab id	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	APAA	CFAM	Phytanic acid
8090	16.8	1.3	-	-	-	0	0	0
8656	21.6	0.9	-	-	-	0	0	0
8890	11.9	1.1	-	-	-	0	0	0
8900	13.2	1.9	-	-	-	0	0	0
9086	11.6	1.1	-	-	-	0	0	0
9092	2656.5	0.9	-26.8	-27.7	-0.9	1	1	0
9189	792.9	1.0	-26.7	-27.8	-1.1	0	0	0
9296	1134.6	0.7	-	-	-	0	0	1
9343	1101.2	0.7	-	-	-	0	0	0
9359	458.4	0.8	-	-	-	0	0	0
9409	858.6	1.5	-	-	-	0	0	1
9470	892.9	0.7	-	-	-	1	1	1
9565	85.2	1.3	-	-	-	0	0	0
9630	1521.0	0.9	-27.4	-27.8	-0.4	0	0	0
9674	1211.1	0.9	-	-	-	1	0	1
9679	2014.8	1.5	-	-	-	0	0	0
9680	499.8	1.0	-27.3	-31.4	-4.0	0	0	1
9684	32.6	1.7	-	-	-	0	0	0
9734	447.6	0.6	-	-	-	0	1	0

Table 26: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses in layer 95. TLE: total lipid extract is expressed in µg·g-1. P/S: Palmitic acid and Stearic acid ratio.

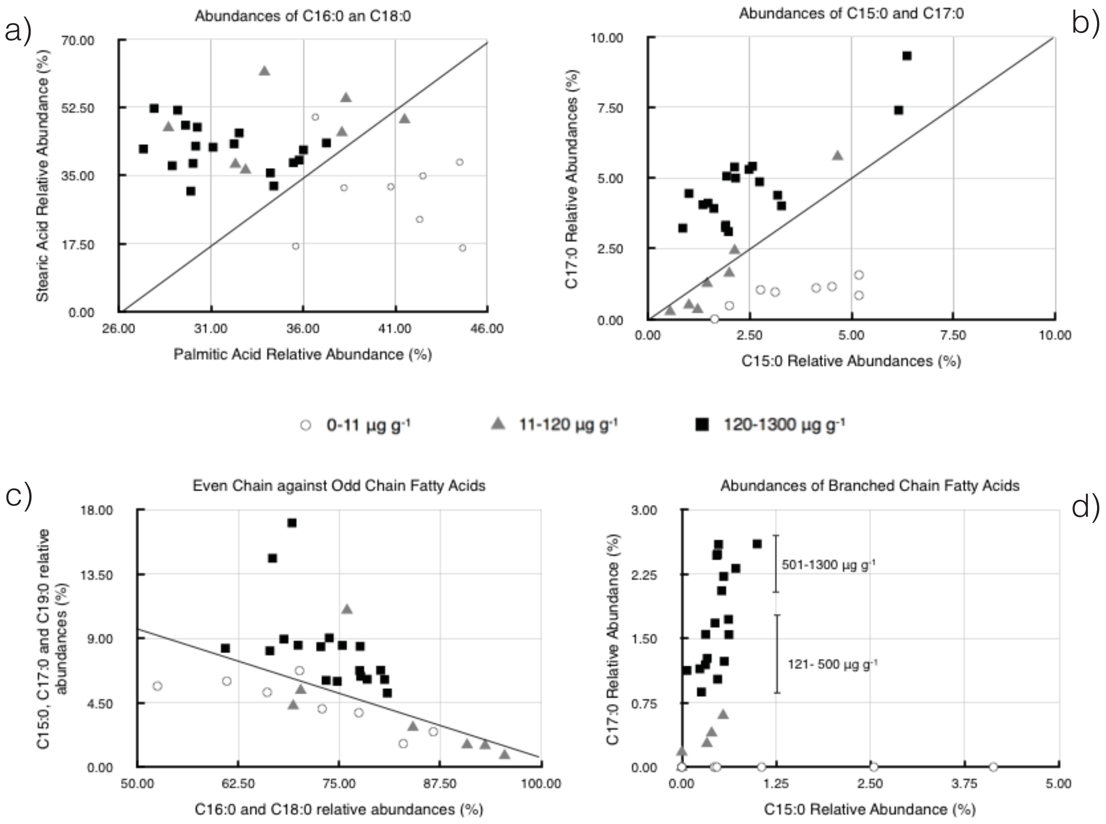


Figure 33: Plot comparing the relative abundances of a) C16:0 and C18:0 b) C17:0 and C15:0, c) odd and even chained fatty acids between 15 and 19 carbons d) C17:0br and C15:0br fatty acids in samples from layer 59.

more abundant in animal tryacylglicerols, plant oils also contain more quantities of unsaturated compounds such as oleic acid (Copley *et al.* 2005a). Moreover, results from the samples under DCM/MeOH extraction have shown the presence of monoacylglycerols and diacylglycerols, which suggests that the source of the free fatty acids most probably are hydrolysed triacylglycerols. Although cholesterol was also detected, the presence of squalene in the samples invalidated it as a biomarker for animal fats. The complete absence of terpenoids, long chain alcohols and ketones that would point to oils and vegetable fats increases the likelihood that the samples did mainly contain animal fat.

In conclusion, 7.7% of the assemblage could have contained non archaeologically significant amounts of fat. In the remaining 92.3%, the samples did not contain sufficient plant or marine biomarkers to suggests these products were significantly present which its free fatty acid profile was not incompatible with the degraded animal triacylglicerides. Comparing phases I and II, it seems that all non-significant residues were detected in stratigraphic unit 59. The fact that all samples in phase I contained significant amounts of lipids might be explained by the difference in sample sizes (95 n= 19, 59 n=32).

Furthermore, a range of specific molecular indicators can be used to explore possible differences between animal fats. One of these is the significant amount of C15:0, C17:0 and C19:0 that some samples present in phase II (Figure 33). The degradation of fats through organically driven β-oxidation is highly common in the rumen. Given that odd-carbon number fatty acids have been found in samples associated with ruminant fats (Evershed *et al.* 2002a), the presence of active rumen bacteria which would also account for the existence of branched chain fatty acids (Dudd *et al.* 1999) and phytanic acid (Ackman and Hooper 1968) should not be rejected. Nevertheless, assignation of a ruminant origin for these fats should be attempted with caution because bacterial activity can also be found in soils. Although they might suggest ruminant fats are not incoherent with the range of detected compounds, these markers alone are not sufficient to establish the probable origin of the animal fats.

One of the more accepted biomarkers for the differentiation between ruminant and non-ruminant fats is the position of the double bond in the chain of the C18:1 fatty acid. As the literature reports (Mottram *et al.* 1999, Regert 2011), bacterial biohydrogenation of dietary fats occurring in the rumen generates a wide variation of double bond positions (Δ^9, Δ^{10}, Δ^{11}, Δ^{13}, Δ^{14}, Δ^{15} and Δ^{16}) in oleic acid whereas non-ruminant fats are largely composed by Δ^9 positions. Nevertheless, the use of this biomarker is only possible when enough quantities C18:1 have been recovered and a dimethyl-disulphide (DMDS) derivatisation is applied. This is not the case of the samples from CRA. Moreover, oxidation degradative processes are particularly efficient in degrading double bonds and byproducts of this oxidation can be found in form of dicarboxylic acids, 9,10-dihydroxy fatty acids, and keto acids (Aillaud 2002, Baeten *et al.* 2010). The chain length of the first ones is indicative of the double bond position of the precursor molecule. Therefore, a wide range of dicarboxylic acids would not imply that the presence of ruminant fats can be rejected (Regert 2011, Regert *et al.* 1998). Again, given that for each precursor, various dicarboxylic acids are known to be formed, their presence with other oxidation products has to be treated with caution. These molecules are the result of the degradation of all unsaturated moieties and, therefore, a certain amount of variation is to be expected.

The vast majority of the samples from CRA present a wide array of dicarboxylic acids and, again, there seems to be a correlation between the amount of recovered lipids and the number of different chain lengths. This fact would add to the non-diagnostic range of evidence for ruminant fats if, as presented, chain lengths were not higher than 17 carbons. Higher numbers would suggest that unsaturated fatty acids with chains longer than 18 carbons were degraded by oxidation. This is the case of many samples in the studied CRA assemblage.

When looking at the relative amounts of each dicarboxylic acid, should oleic acid be the main precursor, azelaic acid could be expected to be the main dicarboxylic acid in the residue. Nevertheless, this is not the case of many archaeological samples, where tridecanedioic acid is the most abundant dicarboxylic acid. The apparent importance of this molecule could be explained by a differential degradation of shorter chains, which would cause the loss of the nonanedioic and decanedioic moieties. Nevertheless, these molecules are usually only detected after acid/base extractions, which accesses compounds more tightly bound with the clay matrix. It is also unclear whether these tighter bond could prevent further degradation. Alternatively, this distribution could suggest the presence of an input of other unsaturated fatty acids given that the range of diacids present would not be incompatible with the existence of longer chained moieties. Paullinic acid (C20:1 cis-13) and Erucic acid (C22:1 cis-13) are both unsaturated fatty acids which could result in an abundance of tridecanedioic acid. The presence of C22:1 in several samples from both stratigraphic units suggests this option should not be completely ruled out.

The presence of unsaturated fatty acids with very long chains has been attributed to residues from marine products (Heron *et al.* 2015). Therefore, the presence of this type of fat sources has to be carefully evaluated. CRA is a site located at the seashore and the recovery of seashells suggests that the sea was being somehow managed, but several extra biomarkers have to be detected before residues from marine products can be securely identified.

Long chain unsaturated fatty acids (C20:1, C22:1 and C24:1), their degradation products, and a series of terpenoids (phytanic acid, pristanic acid and 4,8,12-TMTD) are amongst the biomarkers considered for the presence of marine products. Furthermore, the detection of ω-(o-alkylphenyl)alkanoic acids of 20 and 22 carbons in its chain (Evershed *et al.* 2008a) has been used to argue for the previous existence of polyunsaturated fatty acids. These phenolic molecules are formed by thermal degradation of unsaturated fatty acids which, following a chain of different reactions (see Figure 34), generate benzene rings at the unsaturated positions. Therefore, the creation of these molecules is not solely diagnostic of marine products, but can indicate that the fats inside the vessel were subjected to intense heating (270ºC). In the CRA assemblage, three vessels in phase I (95/9092, 95/9470 and 95/9674) and three more in phase II (59/8726, 59/6405 and 59/6695) contained ω-(o-alkylphenyl)alkanoic acids of 18 carbons

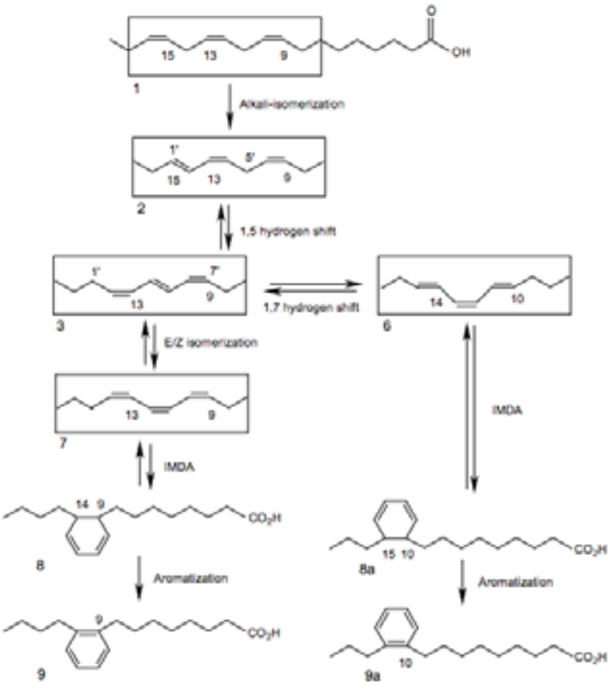


Figure 34: Chain of reactions that transform triunsaturated fatty acids to ω-(o-alkylphenyl)alkanoic acids. Reproduced from Hansel *et al.* 2004.

in the chain (Figure 35). No samples yielded detectable quantities of more than one terpenoid (phytanic acid) and only sherds with high lipid outputs were consistent with the presence of very long chain unsaturated fatty acids. Nonetheless, none of the samples combined all three biomarkers. Therefore, had marine products been contained by vessels in CRA they would have been either degraded to the point they could not be detected, or their quantities could have been so small there was no significant incorporation of their specific markers into the pottery matrix.

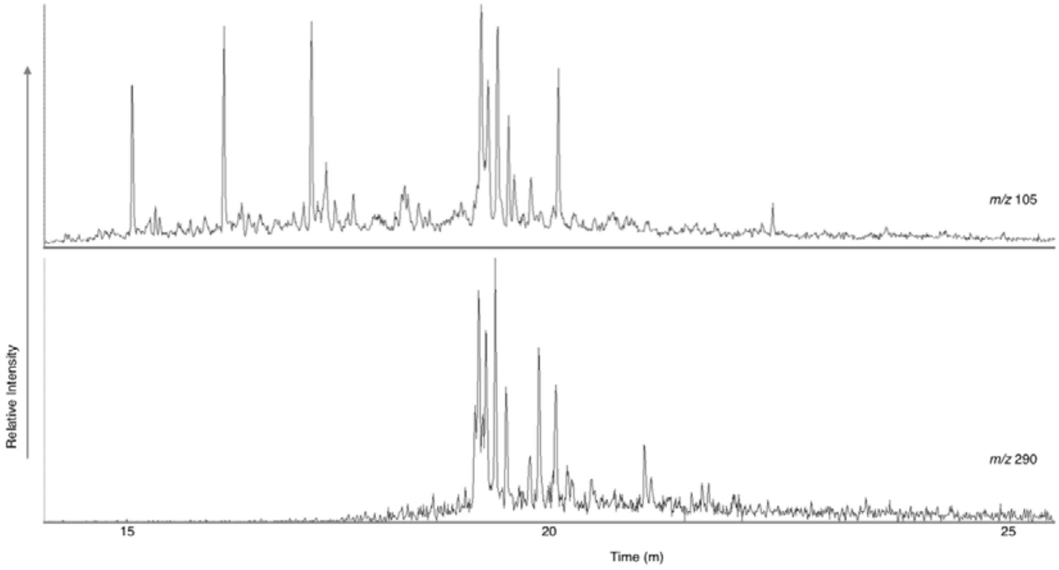


Figure 35: *m/z* 105 and *m/z* 290 mass chromatograms from sample 8726. The 105 ion corresponds to the dialkylbenzene fragment and the 290 is the molecular ion for ω-(o-alkylphenyl)octadecanoic acid.

Although a possible significant input of marine products can be rejected, the presence of ω-(o-alkylphenyl)alkanoic acids does point at the existence of thermally degraded polyunsaturated fatty acids with 18 carbons in its chain. These molecules (linoleic acid and linolenic acid) are present in minor quantities in animal fat triacylglicerides and, therefore, their presence could be expected if said fats had been thermally altered above 250°C. The presence of these temperatures would not be coherent with certain cooking practices such as boiling or stewing, and imply other techniques must not be ruled out. In 5 out of the 6 samples were ω-(o-alkylphenyl)alkanoic acids have been detected, another molecule possibly originating from thermal degradation has also been detected (see Figure 36). Cyclopentyl octadecanoic acids have been shown by Sebedio and others (1989) to be a byproduct of frying oil resulting from similar chemical reactions as the ones followed by ω-(o-alkylphenyl)alkanoic acids but without prior alkali-isomerisation, implying these molecules would not need a clay matrix to undergo cyclization. It has been proposed that cyclopentyl saturated fatty acids would possibly arise from monounsaturated precursors (Dobson *et al.* 1996a and b) but, although experimental and methodological research on archaeological organic residues has previously mentioned the possible existence of these molecules (Evershed *et al.* 2008a), they have not been widely reported in peer-reviewed published case studies.

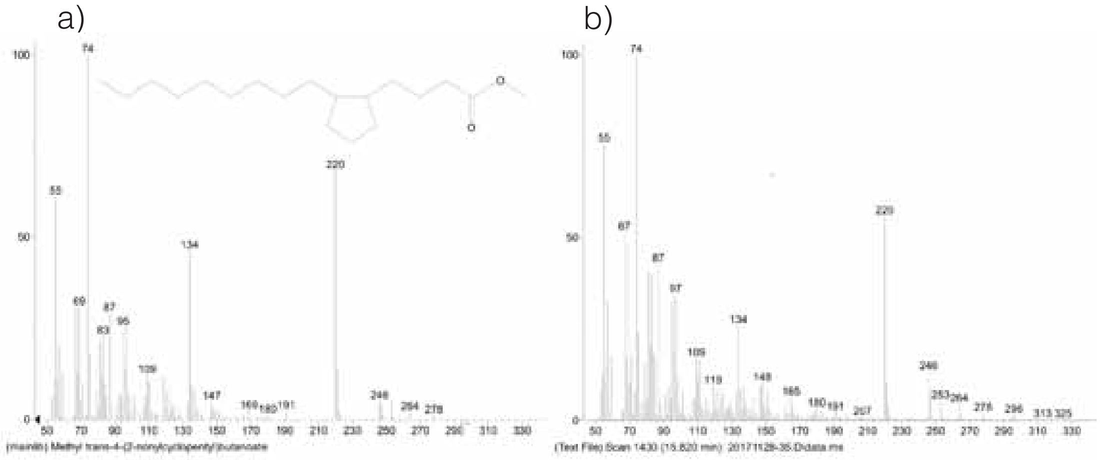


Figure 36: a) spectra of the peak in Rt 15.82 in sample 8726 b) reference spectra of methyl trans-4-(2-nonylcyclopentyl)butanoate, a cyclopentyl saturated fatty acid. (Sebedio *et al.* 1989).

Their appearance together with other markers of thermal alteration provides further support for their origin. Moreover, a cyclopentyl octadecanoic acid is present in another archaeological site within the study (Cova del Vidre, section 6.3), where it is accompanied by a range of mid chain ketones resulting from thermal decarboxylation of saturated fatty acids. Following these results, it should be considered whether other samples presenting only a cyclopentyl octadecanoic acid might have also been subjected to a certain degree of thermal alteration. This would be the case of 95/9734 and 59/8995.

In consequence, the analysis of the recovered biomarkers has been able to differentiate between types of residues in terms of its lipid profile. Nevertheless, it has only been possible to temptatively identify the presence of a fatty acid profile compatible with the degradation of tracylglicerols from animal fats. Although several indicators point that ruminants are amongst the possible animal sources, the high amount degradation could also explain said molecular profiles. The presence of a range of molecules originating from high-temperature reactions indicates that practices submitting these vessels to the aforementioned temperatures can not be ruled out. Furthermore, despite some biomarkers for marine products are present in the assemblage, there is not enough evidence to support their presence. In consequence, the only strategy capable of providing a clearer picture is to obtain the δ^{13}C isotopic values for the palmitic and stearic acids. Research has shown that this technique is able to differentiate between ruminant and non-ruminant fats, fish and terrestrial products, and also between ruminant milk and ruminant adipose fats (Craig *et al.* 2011, Dudd and Evershed 1998, Dudd *et al.* 1999, Evershed *et al.* 1997a, Mottram *et al.* 1999).

Values from the 10 samples (6 from layer 59 and 4 from layer 95) were analysed by GC-C-IRMS and are presented in Figure 37. Results have been plotted together with all other available isotopic data from residues in the Iberian Early Neolithic (Tarifa 2015, Martí *et al.* 2009, Spiteri *et al.* 2016).

This indicator provides correction for the enrichment due to climatic variations (Muckerjee *et al.* 2005) and allows for a clearer interpretation. Initially, samples 59/6231 and 95/9680 fall into the ruminant dairy fat area. The fact that biomarkers for high cooking temperatures have not been found in these vessels could be consistent with the removal of lactose from milk through the application of gentle heat.

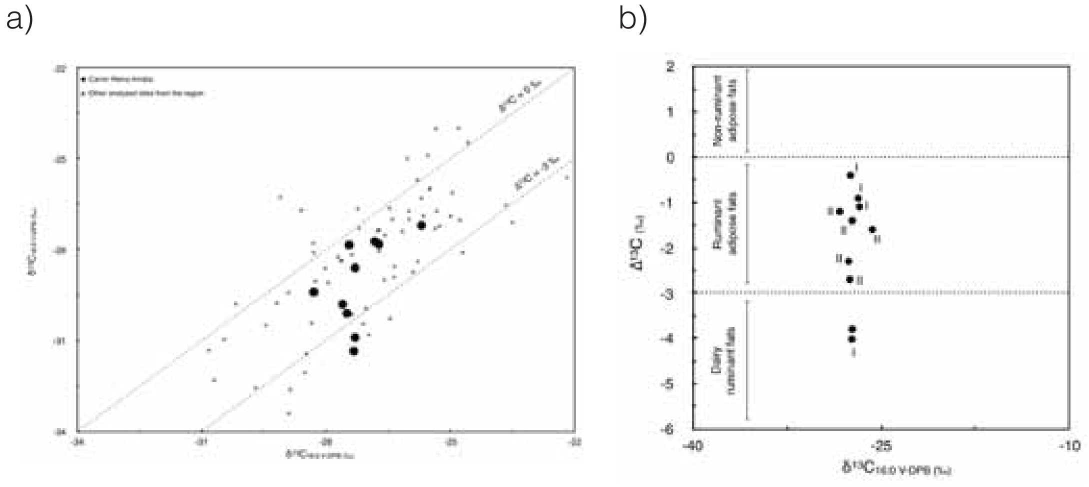


Figure 37: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values. I: Phase I, II: Phase II

Sample 8726 presents the most enriched value. Two explanations can be found for this vessel. The fact that the sample is plotted just in the edge of the marine resources' area could suggest (1) a fish residue. Nevertheless, the absence of other biomarkers that could support this interpretation suggests the presence of (2) a mixture of porcine and ruminant fats. Analyses on Epicardial sites (El Molló, Barranc d'en Fabra and Cova del Sardo) have already reported possible mixtures of ruminant and non-ruminant fats (Tarifa 2015). Therefore, the second hypothesis seems to be the most probable.

The remaining samples plot with the expected values for ruminant adipose fats, which is also consistent with the fact that 78.6% of the faunal assemblage in CRA is composed of ruminant animals.

To better understand the range of uses different types of animals had across phases I and II in Carrer Reina Amàlia, a Bayesian mixing model has been built following a set of assumptions. The absence of significant fish and plant biomarkers has been used as an argument to discard them as possible end members. Therefore, it is assumed that the evaluated samples would be a mixture of unknown amounts of ruminant adipose, ruminant dairy and non-ruminant (most probably porcine) adipose fats. Given that the isotopic signal would result from the repeated use of the vessel to transform and contain products containing a certain amount of fat, the posterior likelihoods for each vessel in the site have been averaged. This procedure has facilitated the obtention of estimates for the importance of each end-member (dairy, ruminant adipose and non-ruminant adipose fats) for phases I and II and for the site overall (Figure 38). Further subtraction of the probability of each percentile of period I from period II has been used to reveal chronological differences.

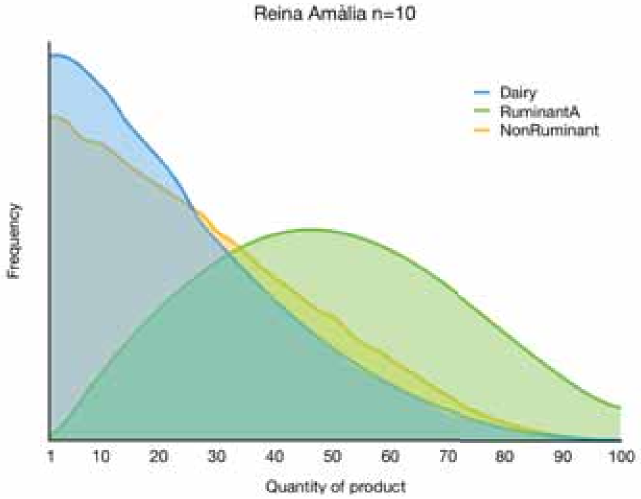


Figure 38: Modelled results averaging all samples isotopically studied in Carrer Reina Amàlia

In phase I (see Figure 39a), the model suggests that ruminant carcass fats would be significantly more prominent than the rest (Dairy median=17%, Ruminant median=47%, Non-ruminant median=25%). As previously seen in the presentation of the isotopic results, only one sample contained a value coherent with a significant input of dairy products. Moreover, the other samples were amongst the most enriched in the assemblage, which the model uses to suggest the possibility that certain amounts of non-ruminant adipose fats could have been mixed with major amounts of ruminant foods during some of the vessel's uses.

In phase II (see Figure 39b), the frequency of uses presents a similar patter to what was observed in phase I (Dairy median=20%, Ruminant median=48%, Non-ruminant median=22%). Although most of the samples have been shown to be coherent with degraded ruminant adipose fats, the model also takes into account a possible minor mixture of dairy with non-ruminant products.

In consequence, the model does not seem to projects any that any differences could be expected between phases I and II. The presence of dairy or mixtures of dairy products with other animal adipose fats may have been more frequent in phase II than in phase I given that more samples have lower $\Delta^{13}C$ values close to the -3‰ threshold (Figure 40). Alternatively, the presence of more enriched and higher $\Delta^{13}C$ values in phase II has been interpreted by the model as a possible higher consumption of non-ruminant products mixed with isotopically enriched ruminant products. Overall, it seems that the culinary practices in Reina Amàlia would have incorporated mostly ruminant followed by dairy fats although the presence of non-ruminant lipids is not rejected by the model.

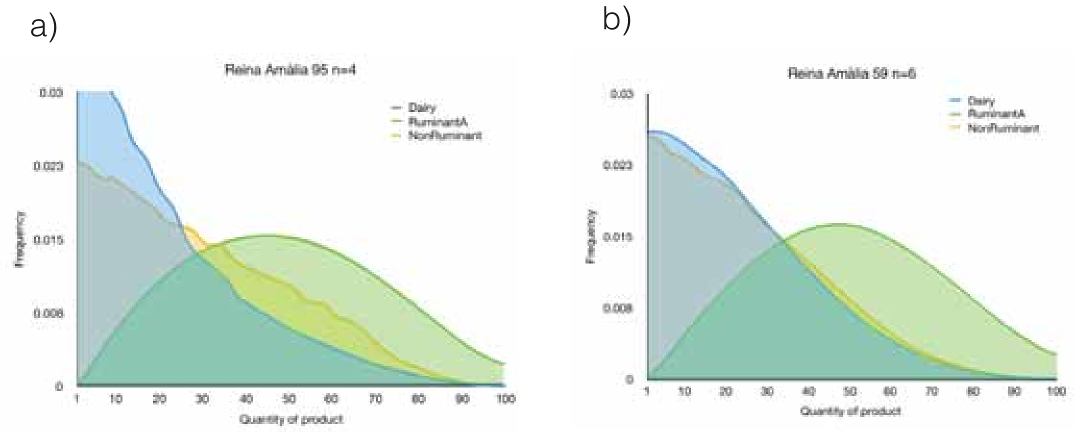


Figure 39: Modelled results averaging all samples isotopically studied in a) phase I and b) phase II.

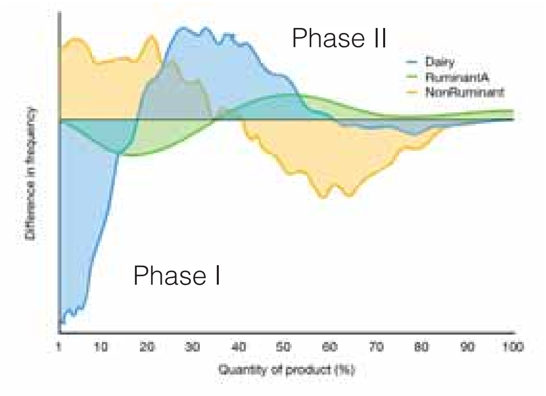


Figure 40: Differences in the statistical distributions between phase I and phase II.

Regarding the possible relationships between shape and contents, previous research has shown that vessel characteristics can sometimes be related to the nature of the content (Mottram *et al.* 1999). In the case of CRA the main molecular indicators for the presence and types of animal fats (TLE, P/S ratio, the presence of ω-(o-alkylphenyl)alkanoic acids or cyclopentyls and phytanic acid) have been compared with the previously presented pottery characteristics (the rim type, the sherd's width, the firing conditions, the decoration and the presence of postdepositional crusts) in phases I and II together. The normality of the data has been tested using the Shapiro-Wilk test and specific parametric or non-parametric statistical tests have been chosen accordingly. The α values were set at 0.05 and p values on the alternative hypothesis (there are no major differences between the analysed variables) are presented in appendix 3.

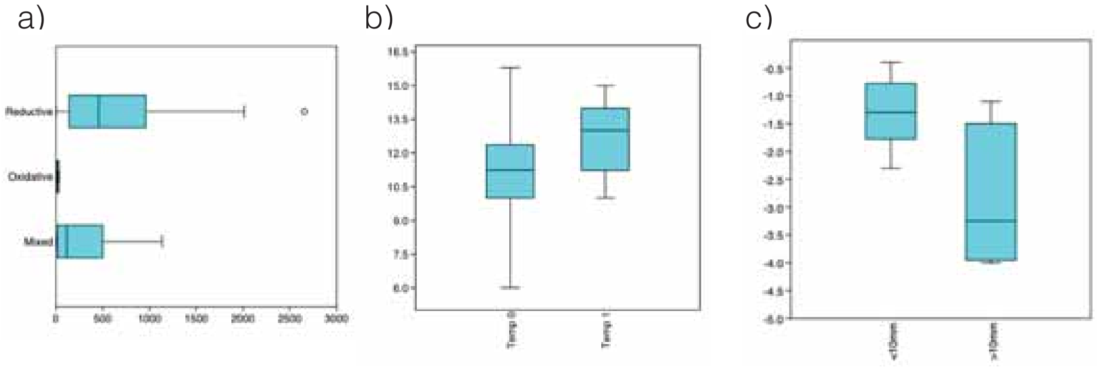


Figure 41: Box plots presenting a) differences in the Total lipid extract ($\mu g \cdot g^{-1}$) as a result of variation in the firing conditions. b) differences in vessel diameters at the rim as a function of the presence of biomarkers for high temperatures. c) differences in $\Delta^{13}C$ values as a function of vessel width being higher or lower than 10mm.

Regarding the concentration of lipids in the sample, it didn't seem that any of the studied parameters was significantly correlated with it. Only in the case of the mainly oxidative firing technique it would seem that lipid concentrations could be lower than the other categories (see Figure 41a). Nevertheless, only four oxidative samples were available and, therefore, these results could be an effect of the small sample size. Neither the rim type nor the presence of handles, appliqués or the absence of plastic applications seemed to be related to any of the molecular biomarkers. In the case of the limestone crusts adhered to some of the studied vessels, it doesn't seem that their presence either helped or compromised fatty acid preservation. Nevertheless, P/S ratios could show a slight tendency towards lower values in samples with no crusts. Given that shorter chained molecules (C16:0) are preferentially leached out due to their higher water solubility, the more degraded the residue, the closer to 0 the P/S ratio would be. Therefore, it should not be rejected that the presence of limestone crusts did partially help with residue preservation as the P/S values tend to be lower in pieces without it. Finally, it seems that the width of the vessel near the rim would be an indicator of some differential practices. It seems that vessels with molecules indicating high temperatures tend to have thicker walls (see Figure 41b), while vessels whose walls are thinner than 10mm were the only ones to contain dairy products or isotopic values which would also be compatible with mixtures of dairy products and ruminant fats (see Figure 41c). The possible correlation between dairy isotopic values and wall thickness has to be interpreted carefully given the small sample size of $\Delta^{13}C$ values below -3‰ (n=2). Moreover, despite biomarkers for the origin of the fat not correlating with any vessel characteristic, the presence of ω-(o-alkylphenyl)alkanoic acids and one cyclopentyl octadecanoic acid seems to be associated with the widest rims.

Finally, given that the spatial situation of the vessels had been recorded, it was possible to assess whether the contents of the vessels could be explained by its specific location inside E-III. Figure 42 presents an interpolation of the TLE in each of the coordinates where the vessel was found. In phase I, the samples with the highest concentration of lipids seem to plot around the same area, while ceramic pieces further away were significantly more depleted (Figure 42a). In phase I, most of the samples closer to structure E-XX, a hearth, seem to present higher lipid concentrations. Nevertheless, another area further from the combustion structure also contained vessels with higher quantities of lipids. The interpretation of the spatial distribution of pottery needs to take into account possible post-depositional effects which would have moved the vessels. Moreover, the fact that they were recovered as sherds instead of complete vessels suggests their positions were not the original ones. In light of this fact, it can be assumed that the random distribution of sherds in the site created a misleading concentration of lipid rich vessels close to the hearth. Alternatively it is also possible that post-depositional effects did not completely disturb the spatial distribution in the site, thus suggesting the higher amount of residues in vessels close to the hearth could be related with the existence of activities related with the preparation and consumption of food.

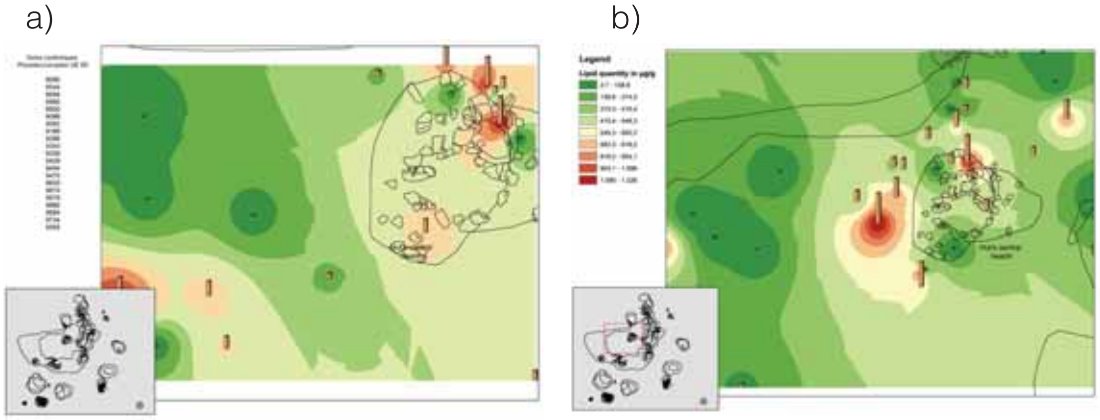


Figure 42: Interpolations representing the abundance of lipid residues (TLE) in vessels located around structure XX, the central hearth of the hut (structure III). a) layer 95 (phase I) , b) layer 59 (phase II).

4.3.6 Conclusion

To conclude, this research has successfully performed 53 analyses of residues from 6400 year old potsherds from a coastal Mediterranean environment. Animal fats where temptatively detected and several indicators such as phytanic acid suggested, amongst other options, the possibility of the fat being of ruminant origin. This assignation was confirmed by the δ^{13}C isotopic values of the C16:0 and C18:0 fatty acids. Although no significant differences were observed between vessel shape and contents, results support three relevant archaeological interpretations:

(1) The confirmation that adipose fat from ruminant animals is widely present in the analysed vessels complies with the fact that the faunal assemblage located in the site is composed of 72% of ruminant species (cattle, sheep and goat) in its NMI. Nevertheless, the consistent presence of ruminant fats implies that consumption of the non-ruminant species is underrepresented. A significant amount of pig (28%) has also been found in E-III, therefore some non-ruminant signals should be expecte.

(2) The presence of ω-(o-alkylphenyl)alkanoic acids and a cyclopentyl octadecanoic acid in eight of the largest and thickest vessels suggests that containers of substantial capacities were used to cook food. Nevertheless, the domestic needs of a nuclear family could have not demanded the use of vessels of such dimensions. In consequence, the practice of cooking for larger groups should be considered. Whether it would be a sporadic communal gathering or a daily practice of shared meals remains unknown.

(3) Apart from the presence of ruminant fat in these samples, milk has been detected in two of the vessels with the thinnest walls of the assemblage. Although the study of the herd management practices in not incompatible with the production of milk (Saña and Navarrete 2016), recent genetic studies concerning the north-eastern Iberian peninsula (Olalde *et al.* 2018 and 2019) suggest that the population could have been highly lactose intolerant. In light of these data, the practice of transformation activities such as the production of fresh cheese should not be discarded. Dairy products could have been used as a complementary source of protein in the diet.

In conclusion, the Carrer Reina Amàlia archaeological site comprises what is today thought to be the oldest build domestic structure in the Barcelona plain. Up to 53 samples from inside the domestic structure were analysed with the aims to identify its possible use and relate it to shape characteristics. The study of the residues indicate that some large vessels were used for cooking with fat from a ruminant origin and the detection of residues from dairy products suggest that the herd management practices might have been more complex than previously anticipated.

5 Garraf massif and Penedès valley

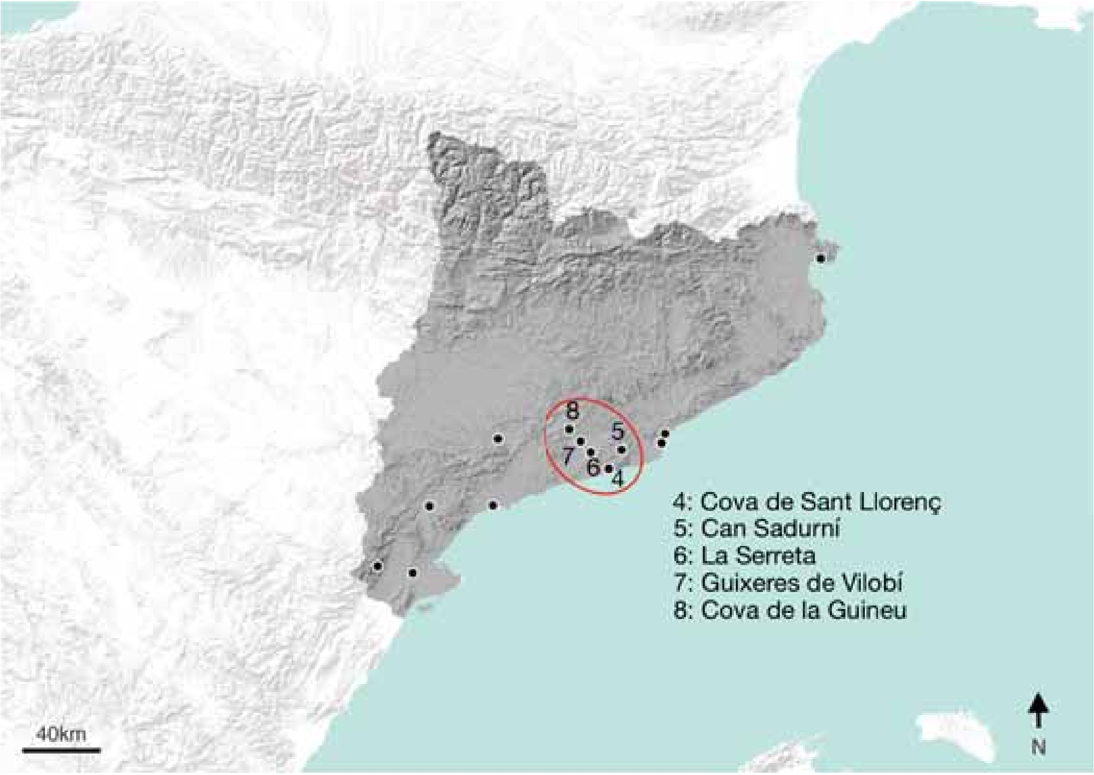


Figure 43: Map of the studied site in the Garraf and Penedès.

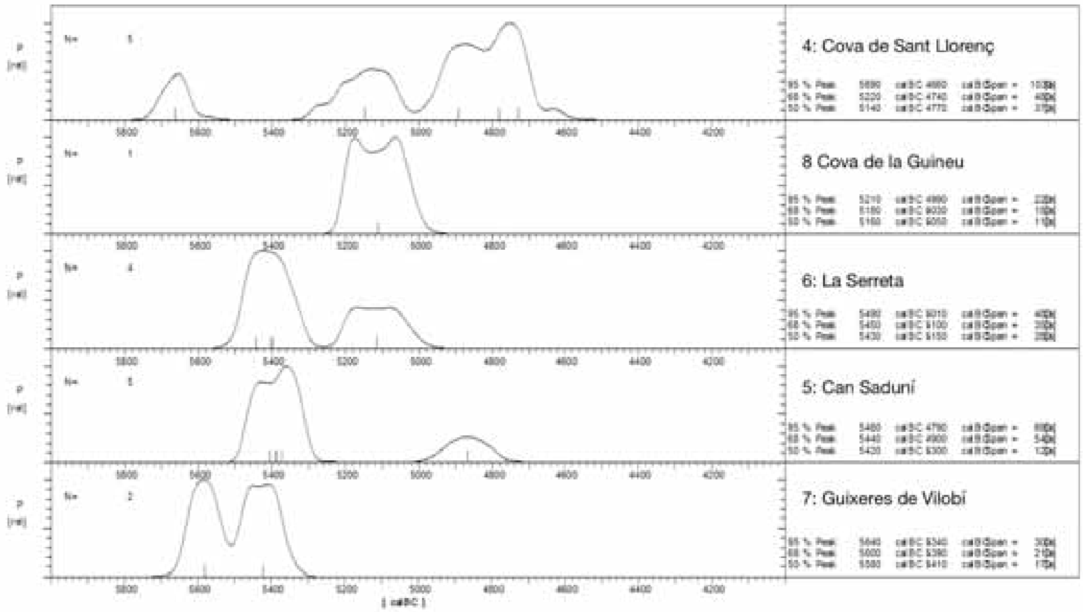


Figure 44: Calibrated ^{14}C dates from Early Neolithic contexts in the Garraf and Penedès. Software: CalPal v1.5

5.1 Cova de Sant Llorenç

5.1.1 Introduction

The Sant Llorenç Cave an archaeological site close to the modern town of Sitges. The cavity used to present a single south-west access leading to a single space, which is around 60m2 wide and measures 8m x 13m in its longest axis. The site is roughly 3km away from the seashore and 245 meters above the sea level. It was formed by a horizontal superposition of different limestone diaclases linked with the process of calcium carbonate solution and precipitation into crusts, stalactites and stalagmites (Borrell *et al.* 2014). Although the modern coastline is different from that of 7000 years ago, the cave is in a privileged setting controlling a sensible range of seashore. Similarly to other caves in this specific position, Cova Sant Llorenç would have easy access to marine products.

The site had been often visited by several explorers such as Miquel Utrillo in 1919, Josep Serra Ciré in the 50's and Miguel Aznar in 1985 when Bellmunt in 1961 performed the first documented archaeological excavation. Records indicate that, by then, sediments were already altered. Before the start of modern excavations in 2007, a wide range of archaeological materials such as cardial pottery, human bones and lithic tools had already been deposited in the Archaeology Museum of Catalonia and the Víctor Balaguer Library-Museum. Between 2007 and 2014, fieldwork performed by the Middle Eastern Prehistoric Archaeological Seminar (SAPPO) research group in the Autonomous University of Barcelona revealed the cave's entire stratigraphy and recovered a complete sequence of in situ archaeological materials below the sediment disturbed by non-scientific excavations (Borrell *et al.* 2011, 2014, 2015, 2016).

5.1.2 Archaeological features relevant in this study

Up to five chronocultural levels (NA) and 6 stratigraphic layers (A) were determined by the end of the excavation. NA1 was placed in pre-Neolithic times while NA2 and NA3 were ascribed to the Early Neolithic. NA4 dated from the Middle Neolithic and NA5 was placed in the Chalcolithic. While level 5 belonged to stratigraphic unit CA4 and level 4 corresponded with unit A6, the three Early Neolithic layers were all part of the same homogenous stratigraphic unit, A4. This was composed mainly of orange oxidised sandy and silty sediment containing limestone blocks rising in number when closer to the unit's base (Borrell *et al.* 2014)(Figure 45).

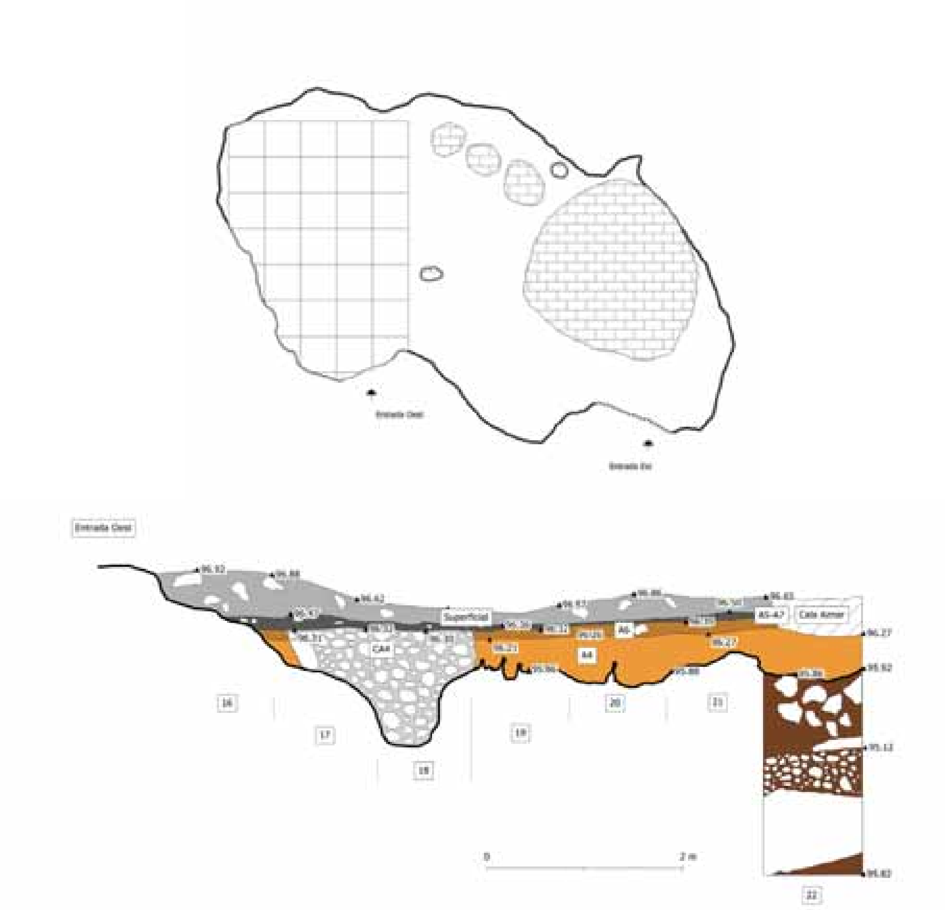


Figure 45: Site plan and section from the excavations between 2007 and 2011. Author: Oriol Vicente (UAB) Source: Borrell *et al.* 2016

In detail, NA1 was the level in contact with the cave's floor. It contained no clear evidence for human occupations although charcoal and animal bones from wild species were recovered in it (Borrell *et al.* 2014). Two Capra pyrenaica bones were ^{14}C dated from 8640±40 BP and 7310±50 BP. respectively, which places this level before the arrival of Neolithic communities in the Iberian Peninsula. NA2 was between 5 and 20 cm deep across the cave and presented archaeological material ascribed to the Cardial Early Neolithic. A bone from Capra pyrenaica from this layer dated from 6750±40 BP. This is a remarkably ancient date which might put this layer right at the beginning of the Neolithic period (Borrell *et al.* 2014) and was later complemented by a Triticum sp. date from 6200±40 BP (Beta 299597) which seems to fit later phases in the Cardial Early Neolithic. Finally, NA3, from 25 to 30 deep, contained 4 negative structures 50cm in diameter and around 30cm deep. A fragmented but well preserved base from a vessel was found in one of these features (E1). Three ^{14}C archaeological dates are available from this phase. The first two (6004±32 BP and 5910±40 BP) are based on domestic ovicaprine bones. The third (5860±40 BP) belongs to a

Triticum aestivum/durum/turgidum charred seed. They are all coherent with a human occupation from the Epicardial Early Neolithic (Borrell *et al.* 2014, Borrell *et al.* 2015).

5.1.3 Archaeological materials

5.1.3.1 Pottery remains

Regarding archaeological remains, the NA2 layer contained middle and big pottery vases, small hemispheric vases and sherds presenting raised handles. Vessel decoration in this layer was performed with plain appliqués and cardial impressions. In NA3, middle sized pottery containers were made using a mould or coils. Surfaces were smoothed and decorations were plain appliqués in orthogonal motives. The base and lower walls of a big container with a possible globular shape were found in one shallow cut. This one (sherd CSL-774) presented a circle shaped appliquée connecting with five radial appliqués placed between the center of the base and the perimeter of this decorative motive (Borrell *et al.* 2014). Furthermore, the vessel's interior contained a carbonised residue which has been analysed.

5.1.3.2 Lithic remains and ornaments

Lithic tools in NA2 where produced by knapping with thermal pre-treatment. Other remains include one stone axe, a piercer on ovicaprid bone and body ornaments (beads) made on seashell. In NA3, remains included one stone axe, flint segments, a sickle blade and a notched piece. (Borrell *et al.* 2014).

5.1.3.3 Animal remains

Animal bones in NA2 belonged to both wild and domestic species such as *Capra pyrenaica*, *Sus scrofa*, *Cervus elaphus*, *Capra hircus*, *Ovis aries*, *Meles meles* and *Vulpes vulpes*. The detailed study of the animal bones is currently underway. In NA3, animal bones contained a lower quantity of wild animals than NA2 and a significant quantity (n=267) of marine malacological remains from the *Palatella caerule*, *Cardium edule* and *Glycimeris* sp. species.

5.1.3.4 Plant remains

According to Antolín (2013), six charred seeds from *Triticum aestivum/durum/turgidum*, *Pinus* sp. and *Medicago* sp. were found in NA2. Layer NA3 contained the highest amount of charred seeds in the site. These were 44 remains from 9 different species including: *Hordeum vulgare*, *Hordeum* sp., *Triticum aestivum/durum/turgidum*, *Triticum diccocum*, *Triticum* sp, *Pistacia lentiscus*, *Prunus avium/mahaleb*, *Pinus* sp. and *Qercus* sp. Other non fully identified remains belonged to the Papilionaceae and Vernonica sp. families (Antolín 2013).

In conclusion, the archaeological remains found in the NA2 and NA3 layers in Cova Sant Llorenç suggest a not intensive but reiterated use of the cavity by Early Neolithic human groups. They would have planned and developed activities in relatively short periods of time (Borrell *et al.* 2016), possibly including the use of the cave as shelter, as a temporary storage place and as a control post, amongst others. Given that no direct evidences of combustion structures were available for the studied period, organic residue analysis on a selection of 4 different vessels was performed to obtain further data on the role pottery could have played in this site. The possible presence of marine products and the existence of molecules indicating significant thermal alterations were considered of special interest.

5.1.4 Sampling and analytical techniques

All selected samples had been found in layer A4. Three vessels were located in the NA3 phase (CSLL14, CSLL15 and CSLL16) while one sample was taken from NA2 (CSLL13). Sample CSLL15 was a visible possible food crust (Table 25). The two selected rims and the sherd with a handle had been fired mainly in an oxidising atmosphere. Furthermore, while sample 2928 (belonging to vessel CSL-774) contained a motive with circular and radial appliqués, sample 1026 was a middle sized container presenting a handle and an initially plain appliquée later decorated with impressions (Borrell *et al.* 2016: Figure 4e).

Lab id	Square	Cut	Id	Shape	Firing	Decoration	Sample weight (mg)	Layer
CSLL13	L19-C	119	2177	Rim	Oxidising	None	100	NA2 Cardial
CSLL14	L21-C	114	-	Rim	Oxidising	None	100	NA3 Epicardial
CSLL15	L21-A	116	2928	Base	Mixed	Circular appliqués	12.4	NA3 Epicardial
CSLL16	M20-D	113	1026	Handle	Oxidising	Impressed appliquée	100	NA3 Epicardial

Table 27: Analysed pottery fragments from Cova de Sant Llorenç

Around 1g of pottery was drilled and lipids were extracted by the acidified methanol extraction. High Temperature Gas Chromatography coupled to Mass Spectrometry (HTGC-MS) was used in all samples to separate and identify specific compounds. Finally samples complying with GC-C-IRMS requisites were submitted for compound specific isotopic analysis. Extractions and GCMS work was performed in the British Museum, Department of Scientific Research and compound specific isotopic analyses were performed in the ICTA-UAB laboratories.

5.1.5 Results and discussion

The samples analysed in Cova de Sant Llorenç presented only minimal amounts of lipids (Table 26). Although 3 out of the 4 vessels contained a significantly high quantity (75%), these values were close to the limit and, therefore, caution must be taken when interpreting its origin. The highest quantity of lipids detected in the site so far originated from the charred visible residue inside vessel CSL774, thus suggesting it could have contained some type of organic product. Samples CSLL14 and CSLL15 contained only minimal amounts of lipids which could be explained by the existence of difficult preservation conditions. Nevertheless, it is also possible that the vessel was either seldomly used to contain and transform fatty rich products or only products with minimal amounts of fats were stored in it.

Lab id	TLE	P/S	Br/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
CSLL13	3.5	1.9	-	-	-	-	Non significant
CSLL14	6.0	3.3	-	-	-	-	Archaeological residue
CSLL15	16.4	2.0	0.016	-	-	-	Degraded animal fat
CSLL16	5.1	2.8	0.023	-23.7	-26.5	-2.9	Degraded ruminant adipose fat

Table 28: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

Regarding the nature of the preserved residues, all samples presented similar profiles of mainly saturated free fatty acids with alkyl chains between 15 and 26 or 30 carbon atoms and a range of unsaturated compounds including C16:1, C18:1 and C22:1. Long chain alkanes and phthalate plasticisers from modern contaminants were equally observed in all samples. Oxidation products such as dicarboxylic acids were detected in sample CSLL15 but no specific biomarkers for high temperatures (long chain ketones, cyclic acids) were found. Only sample CSLL13, which was considered to contain a non-significant amount of residues, presented a peak whose mass spectra could be matched to an unspecified poliaromatic hydrocarbon. Nevertheless, the presence of only one of these compounds is not sufficient to be considered significant. Biomarkers for marine resources (isoprenoic faytty acids, very long chain unsaturated fatty acids, and ω-(o-alkylphenyl)alkanoic acids) were equally absent and no diagnostic molecules for plant organic matter (specific sterols, odd over even alkanes, alcohols and wax esters) were detected either.

Given these results, it must be taken into account that the detected range of free fatty acids in two of the three significant samples is not incompatible with the degradation of animal fat triacylglycerides. Nevertheless, this is not the case of sample CSLL14. It remains unclear wether the presence of more than three times as much palmitic than stearic acids is also compatible with plant triacylglycerides. Even so, the absence of any further markers makes the interpretation of this residue challenging and, therefore, it has been considered only a residue most probably originating from vessel use. The relative amount of C17 branched chained fatty acids compared with the amount of C18:0 from CSLL15 and CSLL16 is coherent with other possible animal fats found in the study but not specific of either ruminant or non ruminant fats. To obtain more data capable to resolve differences between animal fats, in the case of sample CSL16, the presence of significant residues coupled with the absence of problematic contaminants motivated its further analysis under GC-C-IRMS to obtain compound-specific $\delta^{13}C$ isotopic values.

Initially, CSLL16 presents isotopic values not coherent with any modern reference non-ruminant fats (Figure 46). While these can be rejected, given that $\Delta^{13}C$ value is close but not below 3 the absence of dairy products is more difficult to ascertain. Nevertheless, although some dairy reference samples might present similar $\Delta^{13}C$ values, so do domestic and wild ruminant adipose fats. Furthermore, $\delta^{13}C16:0$ absolute values below -25‰ indicate this fatty acid is significantly enriched. In the absence of any C4 plants in the carpological studies and the presence of a significant quantity of marine products in the site, the possible partial input of marine fats could be proposed as an alternative explanation, but only if supported by other biomarkers. When taking into account the C17:0br/C18:0 ratio, CSLL16 shows a value more coherent with the presence of ruminant adipose fats than non-ruminant. Moreover, the absence of any biomolecular indicators for the presence of marine fats suggests that, should there have been any mixture of these two products, only minor amounts of aquatic resources would have been used. These would be coherent with the fact that trace amounts of isoprenoic and polyunsaturated fatty acids could have been easily oxidised and, when no high temperatures transform said molecules in cyclic acids, lost.

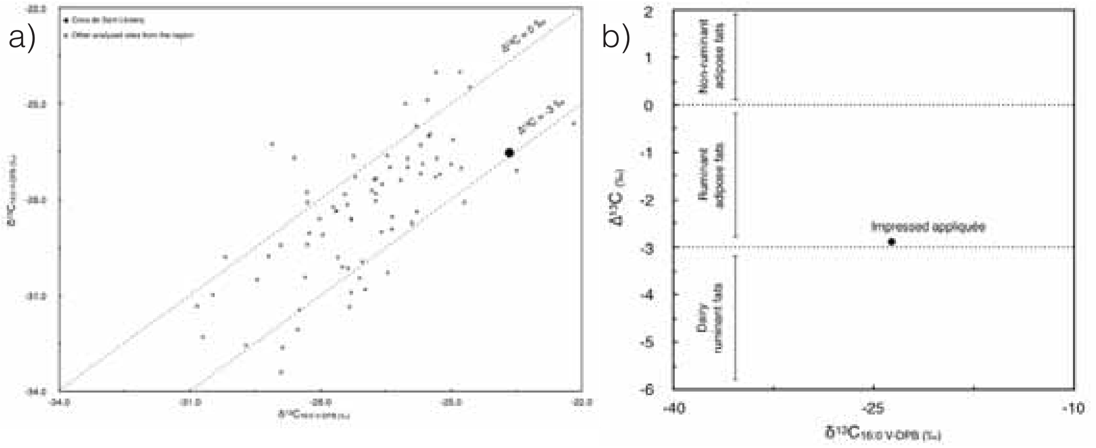


Figure 46: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

To further evaluate possible mixtures a bayesian mixing model including dairy, ruminant and non-ruminant end members has been applied to sample CSLL16. Results are expressed as the probability for the existence of each quantity of product in the mix.

The range of probabilities obtained by the model suggest that dairy products could have been the main content of the vessel (median=43%) while a significant presence of non-ruminants (median=24%) and/or ruminant adipose fats (median=18%) should not be completely ruled out (Figure 47). In this case, given that only one sample from Cova de Sant Llorenç could be submitted to compound specific isotopic analysis, it is not possible to extrapolate the results to any trends in the site. In light of this residue and a Δ^{13}C index coherent with ruminant adipose fats being one of the most likely components of the sample, it is not possible to fully reject the presence of dairy products in this site.

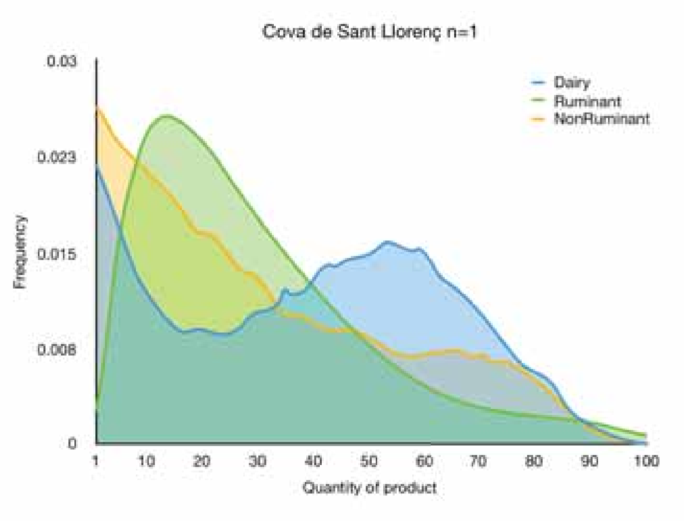


Figure 47: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in sample 16 from Cova de Sant Llorenç.

5.1.6 Conclusion

In conclusion, organic residue analysis in Cova de Sant Llorenç has been able to recover significant amounts of lipids in most of the analysed pottery. Pieces 2928, a base decorated with circular and radial appliqués and, and 1026, a wall with a handle and an impressed appliquée, present a lipid profile coherent with degraded animal fats. The recovered organic remains could have entered the vessel pores after some amount of heating helped mobilise solid fats, although no specific biomarkers for the existence of high temperatures were detected. Nevertheless, the sample with the highest amounts of lipids originated from a visible carbonised amorphous residue in the base of sample 2928. The existence of trace amounts of fat amongst its composition suggests that, rather than a postdepositional event burning the vessels' lasts contents, the residue could originate from repeated everyday use to prepare food. Would this second option be confirmed by further bulk isotopic analyses, the dimensions of this vase would imply large quantities food could have cooked at once.

5.2 Can Sadurní

5.2.1 Introduction

Can Sadurní is a calcareous cave in the Garraf Massif close to the modern town of Begues. The cavity includes an outdoors terrace and an entrance oriented to the south-east; it is roughly 420 meters above the sea level and approximately 15km from the possible position of the Mediterranean coast before the formation of the Llobregat delta (Gámez *et al.* 2011). The nature and position of the cavity has facilitated its constant use by humanity across time. In fact, although its entrance had been apparently closed in 1851 to prevent its use as a shelter, in 1945 the owners of the state reopened it for personal use (Edo *et al.* 2011). Since then, a varied range of excavation activities performed by Joan Bellamunt (Cruz i Ortega 2011), Pere Giró i Romeu and Josep M Masachcs Bolet (Masachcs 1975, Martín i Miret 2011) amongst others retrieved a large set of archaeological materials part of which are now stored in the Archaeology Museum in Barcelona and the Víctor Balaguer Library Museum (Edo *et al.* 2011, Cruz i Ortega 2011). Although some of these remains had been studied by researchers (de la Vega 1972), in 1978, a project leaded by the University of Barcelona started a complete program of exhaustive excavation and documentation (Edo *et al.* 1986, Martín i Miret 2011). Since then, continuous archaeological works in the site have revealed one of the most important stratigraphic sequences for the prehistory of the north-east of the Iberian Peninsula.

The cave's long stratigraphy and the abundance of studies performed on the recovered materials inform about the different activities and economic strategies adopted by the human groups in Can Sadurní. Hiterto, the oldest detected occupations date from the Epipalaeolithic and Mesolithic periods. Afterwards, the site was occupied in the Cardial, Late Cardial and Epicardial Early Neolithic; the Postcardial Neolithic, the Late Neolithic and Chacolithic, the Bronze age and the Iron age (Edo *et al.* 1986).

The Garraf massif is one of the most complex karst systems in the north-eastern Iberian peninsula. As a result, the cavity in Can Sadurní has received the input of exterior sediments as refilling cones from two main points, creating two slopes, one to the north and one to the east. Nevertheless, these features decrease in importance in the most interior areas of the cavity (Edo *et al.* 2011).

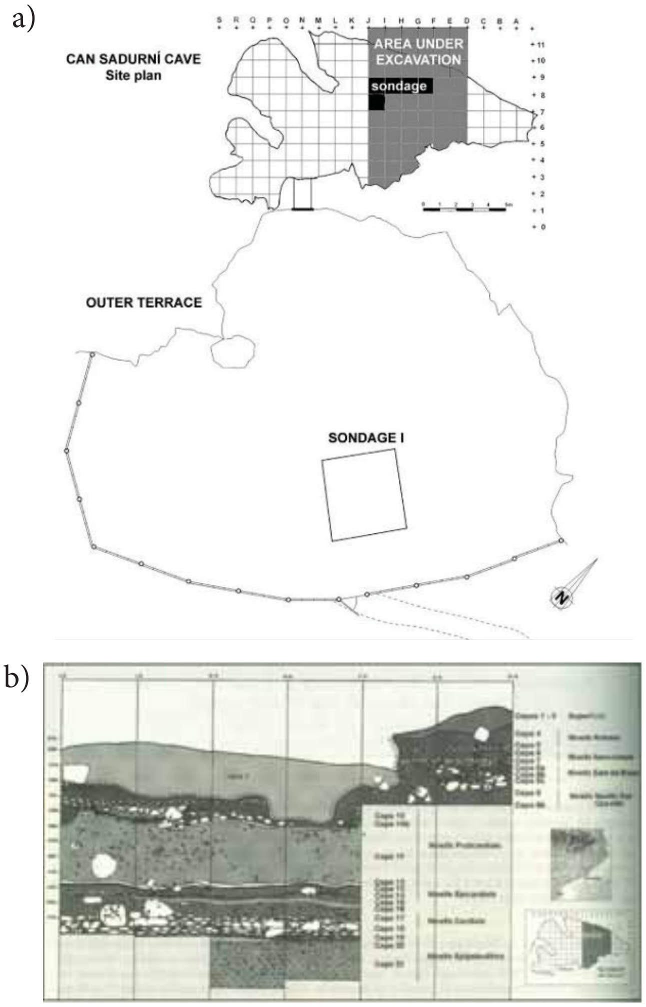


Figure 48: a) Plan of the cave and the outer terrace, reproduced from Antolín *et al.* 2016 b) stratigraphic profile including the layers sampled in this study. Reproduced from Blasco *et al.* 2003

5.2.2 Archaeological features relevant in this study

5.2.2.1 Cardial Early Neolithic (NAC):

Layer 18 is mainly constituted by clayey sediment and big and middle sized blocks originating outside the cave and placed following the slope of the refilling cone (Figure 48). The deposit contained 5 human burials involving female adults and young children with a significant number of grave goods whose location has been linked to packages of seeds that would have possibly been the contents of said containers. The ritual would have involved a primary deposition that was later disturbed by the blocks, thus placing the bones in the secondary position in which they were found. ^{14}C dates from 6375±34, 6391±34, 6405±50, and 6421±34; and specific artefacts such as cardial ware, place this layer in the

Cardial Early Neolithic (Edo *et al.* 2011, 2019). On top, layer 17, between 12 and 14cm deep, presented red to light brown colours and was fully structured and sedimented in a moment of no colluvial activity. Seeds, grinding stones, lithic industry and pottery were also found in this layer. The pottery would still mainly belong to the cardial style, although in a later period, but also included higher percentages of plain appliqués and some incised epicardial motives. In this layer charcoal dated from 6050±110 (Edo *et al.* 2011). At this point in time, a relatively self sufficient human group would have inhabited the cave in a rather permanent way. Researchers propose that the anthropic settlement could have been near the cave cornice while inside could have been used as a place where to dump debris and place an animal shed.

5.2.2.2 Epicardial (NAE):

The epicardial phase in Can Sadurní has been associated with Stratum III, which presented red to brown colours. From top to bottom, layer 12 was whiteish and ashy in its upper part (12a) and organic with debris below (12b). Its irregular distribution presented the usual slope towards the cave's entrance. The soil would have been slightly acidic and possibly associated with a "fumier" episode cleaning the cavity, which would explain the scarcity of archaeological material in the last Epicardial occupation. Immediately below, layer 13 contained grey to brown sediments running 10cm deep with materials coherent with the Epicardial period. Following the stratigraphic sequence, layer 14, 8 to 10 cm deep, presented stones ported by a colluvial event, more abundant archaeological material and charcoal dating from 5980±40BP. Finally, layer 15 contained 7 to 8cm of brown to red sediment with more archaeological remains. This would seem to be the beginning of the Epicardial occupation given that layer 16 is a sterile arrival of sediment, possibly during a period of no human activity in the Cave. Its presence helps better separate these archaeological materials from the lower Cardial set (Edo *et al.* 2011). Following the presence of stabling markers (phytoliths, coproliths and spheroliths) and other indices, the researchers explore the possibility that the interior of the cavity could had been used to shed the herds (Blasco *et al.* 1999). This, a working hypothesis, will be further evaluated when the Epicardial layers are fully excavated. Moreover, the available remains certify the consolidation in all spheres of the Neolithic economic innovations.

5.2.2.3 Postcardial (NP0, NP1a, NP1b, NP2):

Stratum II is composed by a homogeneous sediment with a light brown colour possibly originating from the entrance of soil from outside the cave due to rain and the flow of water. From the oldest to the newest, Layer 11b (NP0) contained big limestone blocks with other middle and small stones. The slope towards the cave's entrance is even more steep than in other times and, in the centre, it became part of layer 11. Charcoal has been dated from 5635±45 BP, 5700±110 BP and 5800±160 BP. In connexion, layer 11 was 71cm deep, it would belong to the Postcardial Neolithic phase 1 (NP1) and it would correspond with Horizon D in the exterior. Charcoal fragments dated from 5350±150, 5470±110 and 5470±140 BP, a sheep preserved in situ dated from 5570±40 BP. Layer 11 has been divided into a number of sublayers, 11a 1-5. Such significant sedimentation preserved an archaeological record indicating that the have had been settled in what has been named the NP1a moment. At this point in time, several finds suggest the human groups in Can Sadurní could he been mining the "Gavà mines" (Bosch and Estrada (coord). 1994) but the use of the cavity would change at the start of what is named the NP1b moment. Then, inhumations were practiced from the cavity's entrance onwards. The dead were placed but not buried and a series of rituals possibly involving feasting (figure 49)(Edo *et al.* 2017) could have taken place. Following the epicardial model, the researchers do not rule out that the funerary area indoors could have coexisted with a human settlement outdoors.

Figure 49: Image of the postcardial burials. Reproduced from Edo *et al.* 2017

At the onset of the NP2 period, big blocks possibly from the collapse of part of the cave's entrance were attributed to layer 10b and dated from 5290±40 and 5340±40 BP. These affected burials from the last phase in layer 11. On top, layer 10, 24cm deep, with brown to yellow colours, would originate in the exterior and present a north-east direction. It would belong to the Middle Postcardial Neolithic and would be contemporary to Horizon C outside the cave. Triticum diccocum from this layer dated from 5279±31 BP (Edo *et al.* 2011). By that time the rise in the total amount of debris and production markers suggest a demographic increase in the group inhabiting the cave. Artefacts suggesting an stronger connection with the Gavà miners and the insertion of the group into wider trade networks might be some of the clues explaining why the cave would be afterwards abandoned and inhabited for a significant period of time (Edo *et al.* 2011).

5.2.2.4 Final Neolithic-Chalcolithic (FN):

At the end of the Neolithic, the cave was used again as a funerary space. Layer 9, around 28cm deep, contained burials from the Late Neolithic and Chalcolithic periods on top a stone bed. Middle Neolithic sediment and materials had been moved to the cave's sides in order to make space for the new burials. Two hearths (structures 2 and 3) were placed between layers 9a and 9b. Structure 2 was dated three times: 4080±100 BP, 4130±110 BP and 4160±160 BP, which fits the archaeological material in this layer, and a human bone from the graves dated from 4225±90 BP. Layer 9b contained small blocks below the burials in layer 9. Two structures have been associated with this layer. Structure 7 was a combustion feature from 4425±50 BP and structure 8 was an accumulation of thermally altered stones which could have been used to heat or maintain food. According to the excavators, the date could be associated with the Veracian phase (Edo *et al.* 2011).

5.2.2.5 Bronze age (BA):

Layer 8, around 28cm deep and with fewer amounts of limestone, presented a significant quantity of charcoal in the top and dated from 2920±100BP. This would belong to the Early, Middle and Late Bronze age. Moreover, layer 8 was subdivided into 8a, 8b and 8c. 8a contained sediment, layer 8b presented small stones probably the soil from 8a and finally 8c with sediment too. 8a mainly belongs to the Late Bronze age and (b and c are mainly ascribed to the Early bronze age) (Edo *et al.* 2011).

In consequence, within the framework of the events between 5500 and 4500, Can Sadurní was the scenario of three different human dynamics. First a funerary use in the Cardial period, secondly a possible settlement during the Epicardial and lastly a place for cattle management at the onset of the Postcardial period. After 4500 the cave would return to its funerary use (Edo *et al.* 2011).

5.2.3 Archaeological materials

5.2.2.1 Early Neolithic pottery remains

In layer 18, a range of different shapes from 33 vessels reveal the presence of six main categories (necked vessels (45.5%), bowls (21.2%), pots (15.2%), jars (12.1%), frustum-shaped vases (3%) and big jars (3%)). Despite this variability, other characteristic shapes from the cardial period were absent, which brought the researchers to conclude some degree of intentionality might have existed in depositing only a subset of the available ceramic assemblage at the time (Blasco *et al.* 2003). Although some shallow vessels were present (21%), the assemblage was mainly composed of deep or very deep vases (79%). Furthermore, recovered pottery sherds were decorated in more than 50% of the cases. These decorations were mainly cardial impressions (80%), plain appliquée (15%) and incised motives (5%). In this layer, the existence of big pottery jars filled with cereal, the presence of specific ovicaprine parts and unused grinding tools suggested the existence of a highly complex ritual (Edo *et al.* 2011).

Layer 17 presented a smaller assemblage where 23.75% of the fragments were decorated (total=215) by lower amounts of cardial impressions (64.71%), higher amounts of plain or decorated appliquée (21.57%) and also higher amounts of incisions (13.75%). Epicardial pottery found in layers 15 to 12 presented an even lower amount (total n=51) of decorated sherds (18.6%) and a significant presence of incised decorations (35.3%) and motives impressed with tools other than a Cardiid seashell (25.7%). Furthermore, curved appliquée and the existence of some specific vessel shapes hinted at the ceramic traditions of the following periods (Edo *et al.* 2011). Finally, layer 11b (NP0) presents an assemblage of Postcardial shapes, surface treatments and decorations which are completely different from previous phases. The Montboló and Molinot styles add up to 83.34% of the assemblage while minor amounts of Cardial (8.32%), Epicardial (5.36%) and plain wares (2.78%) are also present. Although the presence of clear Early Neolithic decorative styles has been explained by the excavations due to the direct contact of layers 18 and 11b, the whole assemblage is still considered to be in transition to the Middle Neolithic. For this reason, it has been placed as the last layer in the Early Neolithic (Edo *et al.* 2011:66).

Regarding the origin of the clays, 34 thin sections from pottery samples in layers 18, 17, 15, 14 and 13 detected the presence of 5 groups of clay types used to produce Early Neolithic pottery (Table 27). Groups 1 and 2 were constituted by granitic rocks found at least 15km away from the cave. Schist in samples from group 3 would indicate this type of clay could have been obtained lower in the Garraf Massif, no more than 10km away from the cave. Finally, groups 4 and 5 could belong to vessels produced using clay from around the archaeological site (Clop *et al.* 2011).

Layer	Group 1	Group 2	Group 3	Group 4	Group 5	Subtotal
Layer 18	10	3	0	0	0	13
Layer 17	4	1	0	0	0	5
Layer 15	1	1	1	1	1	5
Layer 14	4	0	0	1	0	5
Layer 13	5	0	0	0	0	5
Subtotal	24	5	1	2	1	33

Table 29: Groups of clay types detected in pottery from Can Sadurní. Reproduced from Colop *et al.* 2011.

In layers 18 and 17, all analysed sherds belonged to allochthonous clays. This phenomenon would change right at the beginning of the epicardial period with the appearance of vases from local types but in later moments (layers 14 and 13) allochthonous clays would remain dominant. Moreover, anthropic inclusions in the clay for layers 18 and 17 included grog. This is a practice which has been detected in other Cardial Early Neolithic but seems to be absent in layers 15 and 14, where calcite was added to the clay.

In conclusion, although the available published data for most of the Early Neolithic layers is nowadays still based on the 4m2 test trench practiced during the late 90's, the wealth of available data is enough to provide a meaningful pottery assemblage completely coherent with the Cardial horizon in the north-east of the Iberian peninsula. Moreover, thanks to the long stratigraphic sequence, the Can Sadurní cave is a unique witness to the development of new techniques in pottery manufacturing across the Early Neolithic involving changes in decoration, shapes and surface treatments amongst others.

5.2.2.2 Lithic remains

Regarding the lithic industry, a significant amount of jasper and flint was used to produce blades, burins, perforators, scrapers and denticulates. Use wear analysis studied 35 pieces from the Early Neolithic layers. Layer 18 (n=19) presented traces linked with the catchment and processing of animal matter. A small flake was possibly used to discarnate and another fragmented and thermally altered one presented cereal micropolishment in one side. Blade fragments presented wear related to cutting meat or skin and possibly scraping dry skin and two non-retouched blades were used to cut a non-determined soft material. In layer 17 the 16 analysed pieces presented similar results to layer 18. Retouched flakes were used to scrape and cut dry skin and possibly also a non-fibrous plant, and a denticulated flake was used to scrape a non-determined semi-hard product. Blades were used to discarnate to scrape and cut dry skin and a non-retouched blade used to cut a non-determined soft material (Gibaja *et al.* 2011). In conclusion, The studied tools were mainly used to process animal products although two pieces were linked with the transformation of vegetal materials (Gibaja *et al.* 2011).

Microlithic industry remains were not scarce in the Early Neolithic layers with around 41 pieces studied in layer 18, 24 in layer 17 and 21 in the Epicardial period. Nevertheless, the degree of fragmentation was high in the Early Neolithic. Only three complete pieces, including an intensively used grinding hand, could be recovered. Its study by Ache (2011) revealed the presence of grinding stones, axes and plaques involved in the processing of both organic and inorganic products. The bone industry in the layer included two hafting fragments possibly belonging to a bone spoon, a projectile, a triangular piercer and several fragments of spatula. Although these assemblages should be interpreted in a funerary context, they could also belong to other activities not yet fully documented in the cavity in that period (Edo *et al.* 2011).

5.2.2.3 Animal remains

Around 2760 animal bones from the Neolithic period were studied by Saña *et al.* (2015). They showed the presence of *Ovis aries*, *Capra hircus*, *Bos taurus*, *Sus domesticus*, *Cervus elaphus*, *Capraeolus capraeolus*, *Sus scrofa* and *Oryctolagus cuniculus* amongst others. Table 28 presents the percentage of the number of remains for each species across time as they were published by Saña *et al.* (2015). When classifying animal species according to its digestive capacities, table 29 presents the relative quantities of ruminant, non-ruminant and cecotroph remains.

	OVAR	BOTA	SUDO	CEEL	CACA	SUSC	ORCU	DOM
NAC	54.79	2.17	3.98	1.27	1.27	0.54	24.95	71.4
NAE	63.85	4.23	7.51	0.94	0	1.41	3.76	88.05
NP1*	50.2	6.89	14.76	1.57	0.79	1.38	3.94	89.98
NP2	51.48	5.06	10.97	3.38	0.84	0.42	2.11	92.02

Table 30: Percentages of animal remains from the Cardial, Epicardial and Postcardial periods. *:NP0 is included in NP1. Source: Saña *et al.* 2015.

	Ruminant	Non-Ruminant	Cecotroph
NAC	59.5	4.52	24.95
NAE	69.02	8.92	3.76
NP1*	59.45	16.4	3.94
NP2	60.76	11.39	2.11

Table 31: Percentages of ruminant, non-ruminant and cecotroph remains from the Cardial, Epicardial and Postcardial periods in Can Sadurní. Elaborated from Saña *et al.* 2015.

When looking at mortality rates, meat production is deducted from the sacrifice of juvenile animals up to two years after birth while lactic production sacrifices are around 2 months of age. In the case of Can Sadurní, pigs show a clear pattern associated with meat production, which is straightforward given that pig milk was never used as a dietary element. Although the small amount of Bos taurus prevents a clear picture, juveniles were sacrificed across all phases, thus also suggesting a meat production strategy. Domestic sheep and goats were sacrificed at around 2 months of age mainly in the NAC (layers 17 and 18), which is coherent with a dairy production strategy although child mortality could also explain this phenomenon. There is a shift across time towards meat production starting in the NAE (layer 15 and onwards).

Regarding skeletal parts, head bones seem to be overrepresented in domestic taxa while wild animals present an equilibrated amount of remains across the skeleton. This is mainly because of the significant amount of decidual teeth found in the NAC layer, which would point at animal sacrifices during youth. The amount of decidual teeth is significantly reduced (88.5% to 42.6%) between the Cardial and Epicardial phases and remains stable afterwards. Nevertheless, crania would still be overrepresented across the whole sequence. It seems that a significant percentage of the decidual teeth could come as a by-product of stalling. Nevertheless, mortality patterns from the postcranial skeleton suggest this is not the case for the NAC, but could be a significant phenomenon in the NAE and NP periods (Saña *et al.* 2015).

Regarding foddering strategies, a significant abundance of charred fruits from *Arbutus unedo*, *Pistacia lentiscus*, *Quercus* sp. and *Vitis* sp. seeds appear in the NP periods. It should not be ruled out that these plants could have been used as animal fodder. To obtain more direct data on pig foddering strategies, studies by Navarrete *et al.* (2017) evaluated the $\delta^{13}C$ and $\delta^{15}N$ isotopic values in pig bone collagen from layers 10 (n=2), 11b (n=1) and 17 (n=1) in Can Sadurní. Although the number of specimens analysed (n=4) is not high, they are significantly enriched in both $\delta^{13}C$ and $\delta^{15}N$ compared with other sites such as La Draga and Cova del Frare. The $\delta^{15}N$ values could suggest that a relatively high proportion of animal sources contributed to the proteins ingested by both domestic and wild pigs. For Can Sadurní, this would also be coherent with the archaeobotanical and zooarchaeological analyses, which suggest a home-based management of the heard. Furthermore, this interpretation is also similar to the one for pig management in Caserna de Sant Pau. Although these analyses were performed on collagen, it is to be expected that other tissues would have been affected by the animal diet as well.

Regarding marine remains, most probably post depositional effects severely altered the exterior of seashells. In the case of these decalcified malacological remains, it would seem impossible to specify the species, but the researchers suggest their main use was ornamental and ritual. Two seashell bracelet fragments and polished and non-perforated seashell appliqués, which could have been part of a textile piece, were amongst the recovered artefacts (Blasco *et al.* 2003).

5.2.2.4 Plant remains

Layer 18 contained up to 49332 carpological remains. This is an exceptional and unique assemblage across the catalan coast. Moreover, between 3000 and 6000 remains/litre were found inside pots, which would suggest the last contents were still present in the vessels at the moment of the excavation. The study of the seeds revealed a mixture of different wheat and barely species inside. They had been completely processed before the deposition as grave goods but they were most probably charred inside the vessels. The heating treatment would be around 250 to 300 degrees. Although weeds and ruderals were detected in the assemblage, none were found inside pottery. Wild taxa in this layer would have been most probably incorporated to the cave as fuel (Antolín, 2013).

Layer 11 contained a wider botanical diversity than layer 18 although in this case 1814 botanic remains were recovered. These now included legumes and seeds that were interpreted as debris from everyday cooking. The formation of this assemblage could have involved several human actions, an idea which is reinforced by the fact that there is an area richer in *Arbutus unedo* and another with a more significant amount of cereals. It has to be taken into account that, differently from layer 18, the creation of layer 11 would have been a much slower process. Evidences of beer production in pottery and grinding stones were found in this layer (Antolín, 2013).

Layer 10 shared many similarities with layer 11. The 229 recovered remains would include peas and lentils which would have been charred between 300 and 350ºC. Wild remains for this one and layer 11 could have been possibly used as fodder for the animals during winter months (Antolín, 2013).

Regarding recovered charcoal fragments, studies by Piqué (Blasco *et al.* 1999) and Ros (Edo *et al.* 1999) indicated the existence during the Early Neolithic of a litoral thermophilic maquis with *Quercus caducifolis* (oak), *Chamaerops humilis* (Mediterranean dwarf palm), *Rhamnus alaternus* (Mediterranean buckthorn), *Pistacia lentiscus* (lentisk), *Arbutus unedo* (strawberry tree), and *Olea oleaster* (wild olive tree) combined with other submediterranean species. Furthermore, it would seem that a higher amount of deciduous oaks and the presence of species from more humid environments such as Prunus or Rosaceae would be slowly substituted as the Neolithic advanced by a more thermophyllic and heliophillic landscape also including Erica sp. and conifers such as Pinus halepensis, and Juniperus sp.

5.2.2.5 Organic residues

Can Sadurní was the first Neolithic site in the Catalan coast in which residue analyses from across the stratigraphic sequence had been published before this study (Blasco *et al.* 2008, Spiteri 2012, Spiteri *et al.* 2016). The first analyses were performed by Treserras and Matamala and included starch and phytolith analyses, extraction of lipids and spot tests for specific products (Tresserras 1997a).

In these works, 10 fragments from layer 8a were analysed and 5 presented significant results. Two other residues on fragments of flat bases were interpreted as originating from dairy products and finally a residue from a vessel wall was attributed to degraded animal fats. Although the PhD thesis from Treserras (1997a) details lipid analyses were performed after soxhlet extractions, the absence of GC-C-IRMS data to validate the presence of dairy products suggests results must be treated with extreme caution given that dairy products have been previously mistakenly identified by sole detection of very short chain fatty acids. Although fatty acids with chains shorter than 12 carbon atoms are found in higher quantities in milk triacylglycerols, degradation factors can fracture longer chain moieties or completely leach original molecules. The use of $\delta^{13}C$ values of palmitic and stearic acids is the nowadays most accepted strategy to validate the presence of said product. Finally, two big flat based jars in the early Bronze age layers presented traces of beer (individuals 10 and 105). This drink was identified following the presence of gelatinised starch which showed symptoms of an enzymatic attack, yeast evidences, phytoliths and silica skeletons some of which belonged to barely and diatom frustules. No calcium oxalates were detected in the Bronze age samples (Blasco *et al.* 2008).

One pottery vessel from the postcardial layers was also analysed. This showed the presence of calcium oxalate and some phytoliths, which was interpreted as evidence of fermented beer. Starch in two grinding stones presenting malting indices was also reported as supporting evidence for the presence of locally produced beer (Blasco *et al.* 2008).

As reported in her PhD (Spiteri 2012) and following articles (Spiteri *et al.* 2016), Cynthianne Spiteri performed organic residue analyses on 31 ceramic vessels from layers 18 (n=12), 17 (n=15 and 1 soil sample), 13 (n=1) and 10 (n=1). Residues were recovered using a DCM/MeOH extraction and positive results were obtained in 42% of the vessels (n=13). The hydrolysis products of triacylglicerols were identified in several samples and long-chain ketones were present in five vessels accompanied, in one case, by ω–(o-alkylphenyl) octadecanoic acid isomers. Wax esters were detected in two samples along with very long chain fatty acids and alcohol doublets, indicating the degradation of the wax esters into its constituents. Therefore, in these cases the presence of plant products could not be rejected.

In layer 18, only 3 out of the 12 vessels retained significant amounts of fat (25%). Ranges of alkanes, alcohols and wax esters were observed in the residues from this layer. Samples 01 and 13 were submitted to GC-C-IRMS analyses revealing the presence of a ruminant adipose fat and a ruminant dairy product. The low amount of lipids recovered for the main cardial phase was used to suggest that the negligible amounts could be indicative of the storage and processing of plant materials (Spiteri 2012). Furthermore, some of the molecules recovered also suggested the existence of a plant input. In layer 17, 8 out of 15 vessels presented significant amounts of fat (53%). Possible plant inputs were detected in some of the vessels, one of which had a P/S ratio of 5. Isotopic analyses revealed that three samples contained dairy fats and an extra one was attributed to ruminant adipose fat. The researchers further reported that only one sample from layers 13 and 10 had provided a significant residue. It is significant to note that, after this study, no animal fats coherent with porcine products had been reported. This could be coherent with the fact that pig bones were less than 5% in the Cardial Early Neolithic assemblage.

Complementing Spiteri's research, this study aimed at incorporating samples from across the stratigraphic sequence. Three new samples from layers 17 and 18, three samples from layers 12 to 14 and one sample from layer 11b were selected to enrich and validate the picture obtained in previous studies. Furthermore, samples were selected for the NP1a and b phases (n=2 and n=4 respectively) and three more ceramic pieces were selected from layers 10 and 10b, which would have corresponded with the NP2 phase. Beyond the Postcardial Neolithic and complementing the work by Tresserras and Matamala, two samples from the final Neolithic (layer 9b) and three sherds from the Bronze Age (layer 8) were incorporated in the study as well. Although the focus of this research is to better understand the uses pottery had in the Early Neolithic, in the case of Can Sadurní, data across the sequence has been used to better inform the exceptionality and importance of the available data between 5500 and 4500 cal BC.

5.2.4 Sampling and analytical techniques

Lab Id	Layer	Square	Arch idw	Arch phase	Shape	Firing	Decoration	Sample weight
8	17/18	I8	69-97	NAC	Rim	Oxidising	None	0.99
9	18	I7	50	NAC	Wall	Oxidising	Cardial	1.02
12	18	I7	2	NAC	Rim	Mixed	None	1.02
1	14	H8	65-63	NAE	Rim	Mixed	None	1
10	13	I8	45	NAE	Rim	Mixed	Handle	1.03
11	12	H8	21	NAE	Wall	Oxidising	Incised appliquée	0.99
6	11b	I5	1	NP0	Rim	Mixed	Plain appliquée	1.1

Table 32: Analysed pottery fragments dated between 5500 and 4500 (n=7) (layers 12 to 18).

The seven analysed samples were mainly rims (71.4%) and two walls (28.6%) presenting either oxidised or mixed firing techniques (Table 30). Amongst the cardial samples, two were non decorated and one presented seashell impressions. In the Epicardial and Postcardial phase 0, two samples had appliqués while the other two were deprived of any specific decoration (the handle in sample 10 was interpreted as a functional feature).

Lab Id	Layer	Square	Sherd	Phase	Shape	Firing	Decoration	Sample Weight (mg)
4	11	H4	27	NP1a	Rim	Mixed	Orth appliquée	100
2	11a4	E7	92	NP1a	Rim	Mixed	Nipple	105
13	11a1	I10	64	NP1b	Rim	Reducing	None	106
7	11a2	D10	58	NP1b	Rim	Oxidised	Plain appliqués	114
3	11a3	G10	8	NP1b	Rim	Mixed	None	104
20	11a3	E10	60	NP1b	Wall	Reducing	None	104
21	10b	E11	204	NP2	Rim	Mixed	Montboló handle	99
14	10b	E9	26	NP2	Rim	Reducing	None	1
5	10b	D7	6	NP2	Wall	Mixed	Tab	111
16	9b	F10	19	FN	Rim	Mixed	None	1
17	9b	H8	28	FN	Rim	Reducing	Tab	1
15	Ie9	G10	14	BA	Rim	Oxidised	None	101
18	I89	F11	12	BA	Rim	Mixed	None	102
19	8a	E8	26/6	BA	Rim	Reducing	None	104

Table 33: Analysed pottery fragments from occupation phases after 4500 cal BC (n=14).

Similarly, the 14 samples located in layers after 4500 were mainly rims (85.7%) and two walls (14.3%). The studied sherds presented a range of characteristic features depending on their layer of origin (Table 31). While 8 samples contained no specific features, the remaining six had a range of plastic applications such as nipples, plain appliqués, tabs, and a Montboló style handle.

Samples were extracted via acidified methanol and submitted to High Temperature Gas Chromatography coupled with Mass Spectrometry (HTGC-MS) in the laboratories in the British Museum. Further compound specific analyses of palmitic and stearic acids was carried out at the ICTA laboratories in the Autonomous University of Barcelona.

5.2.5 Results and discussion

All samples from the Early Neolithic layers seemed to contain lipids above the $5\mu g \cdot g^{-1}$ threshold (Table 32), which could indicate, in this case, a preservation rate of 100%. Sample CS10 contained the second highest amount of residues recovered in the site (4.8mg). These are quantities which have previously been reported in contexts with good organic matter preservation (Correa-Ascensio and Evershed 2014). Other samples such as CS9 and CS12 equally presented significant quantities of lipids. It would seem that, in this case, despite the samples would have been excavated between 1997 and 2013, organic residues would still be present in high amounts. Nevertheless, contaminants such as phthalic acids were equally detected. Previous data obtained by Spiteri (2012) performing DCM/MeOH extractions and the fact that, in one case, even after the stablished acidified methanol extraction, wax esters could still be detected in the chromatogram (CS20), suggests solvent extractions on selected samples could help provide data on more complex and informative molecules.

Lab id	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
CS1	87.2	1.1	-	-	-	Degraded animal fat
CS6	35.7	1.2	-	-	-	Degraded animal fat
CS8	49.9	1.0	-	-	-	Degraded animal fat
CS9	188.6	1.5	-26.0	-26.7	-0.7	Degraded ruminant adipose fat, thermal alteration
CS10	4832.3	0.8	-27.0	-30.8	-3.8	Degraded dairy ruminant fat, thermal alteration
CS11	21.4	1.2	-	-	-	Degraded animal fat
CS12	261.8	3.6	-24.6	-24.5	0.1	Degraded non-ruminant animal fat thermal alteration

Table 34: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses in samples between layers 12 and 18. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

The quantities of lipids do not seem to vary significantly between samples before and after 4500 cal BC. While the highest and the lowest amounts of residues were found in the Early Neolithic layers, samples from the NP1 to the BA phases were equally rich in lipids and waxes (CS5=3.5mg, CS14=3.98mg, CS20=6.6mg). Across all chronological phases, oxidation products such as long ranges of dicarboxylic acids and different keto octadecanoic acids could have resulted from the oxidation of mainly oleic and palmitoleic acids (amongst others).

Most of the analysed residues from the Early Neolithic layers present a fatty acid profile not coherent with the degradation of plant triacylglycerols. Moreover, the absence of sitosterol and the presence of cholesterol and a possible cholesterol derivative (cholesta-3,5-diene) in samples CS9 and CS11 might suggest the presence of fatty acids resulting from the hydrolysis of animal triglycerides. This interpretation is supported by the fact that the P/S ratio is not incoherent with said degradation. No isoprenoid acids, C20:0 or C22:0 ω-(o-alkylphenyl) alkanoic acids, very long chain unsaturated fatty acids or their dihydroxy oxidation products were detected in any of the samples, which prevents the identification of an input of marine matter.

Lab id	TLE	P/S	Molecular markers	Interpretation
CS2	61.6	1.29	Phytanic acid, D9-16,	Degraded animal fat
CS3	39.55	0.99	D9-D13, Cholesterol	Degraded animal fat
CS4	63.3	1.92	D9-D11, Cholesterol	Degraded animal fat
CS5	3458.6	0.55	ω-(o-alkylphenyl) octadecanoic acid, long chain ketones, D9-D17, Ricinoleic acid (traces), 2-hydroxy 16 to 18:0	Degraded and heated animal fat, possible plant imput
CS7	1373	0.67	Phytanic acid, D9-18, Cholesterol	Degraded animal fat
CS13	91.9	0.92	D9-D12, Cholesterol	Degraded animal fat
CS14	3985.6	1.07	D9-D18, C24-27 alcohol, 15-hydroxy 16:0, 17-hydroxy 18:0, 19-hydroxy 20:0, 21-hydroxy 22:0, 23-hydroxy 24:0 25 hydroxy 26:0, 14-hydroxy 16:0, 16-hydroxy 18:0, 18-hydroxy 20:0, ω-1 diols (1,23 C24, 1,25 C26, 1,27 C28 1,29 C30), fatty alcohols (24-27), cholesterol	Degraded animal fat with possible beeswax input
CS15	38.1	2.3	D9-D11, Cholesterol	Degraded animal fat
CS16	518.1	1.48	Phytanic acid, D9-15	Degraded animal fat
CS17	41.9	1.38	D9	Degraded animal fat
CS18	64	1.9	D9-D12	Degraded animal fat
CS19	74	0.76	None	Degraded animal fat
CS20	6617	1.37	D9-18, 15-hydroxy 16:0, , 17-hydroxy 18:0, 19-hydroxy 20:0, 21-hydroxy 22:0, 23-hydroxy 24:0 25 hydroxy 26:0, 14-hydroxy 16:0, 16-hudroxy 18:0, 18-hydroxy 20:0, ω-1 diols (1,23 C24, 1,25 C26, 1,27 C28 1,29 C30), 24-29 OH, wax esters (traces)	Possible wax
CS21	353.8	0.79	D9-18, Cholesterol	Degraded animal fat

Table 35: Results from the lipid extractions and the GC-MS analyses from samples above layer 12. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

Regarding layer 18, recent interdisciplinary research revealed the presence of evidences of fire on multiple archaeological remains. Charcoal studies revealed the presence of radial cracks associated with the combustion of green wood, thermal alterations were detected in both human (mainly cranium fragments) and animal bones (no more than 30% of the assemblage) while lithic and macrolithic tools also presented evidence of exposure to high temperatures. Moreover, the significant charred seed assemblage found in this layer presented signs of alteration associated with their containers, the pottery, but did not show any evidence of germination or insect attack, which would suggest fires could have affected the assemblage only shortly after its deposition. This evidence was seen by the researchers as coherent with the possible existence of the inclusion of fire in at least some of the funerary rites practiced in Can Sadurní during the

Early Neolithic (Antolín *et al.* 2011, Edo *et al.* 2019). The recovered residues from Spiteri (2012) and the results detected in this study are in complete agreement with this possibility. The presence of long chain ketones from free fatty acids interpreted as the result of a ketonic thermal decarboxylation in the samples analysed by Spiteri (2012) is complemented by the results obtained from samples CS9 and CS12, which show the presence of ω-(o-alkylphenyl) octadecanoic acids, a cyclopentyl octadecanoic acid and the possible existence of trace amounts of the K31 long chain ketone.

Previous micromorphology studies suggested the bone and charcoal detected microremains would have not been exposed to temperatures above 300ºC and the study of the lithic tools also proposes the existence of a temperature no higher than 350ºC. Complementing this data, the range of molecules detected in the residues can only be created above certain energy thresholds. Following experimental studies (Evershed *et al.* 1995a, Raven *et al.* 1997, Hansel *et al.* 2004, Evershed *et al.* 2008a) both ω-(o-alkylphenyl) octadecanoic acids or long chain ketones have been shown to appear after exposure to high temperatures. While these are usually above 200ºC for ω-(o-alkylphenyl) octadecanoic acids, studies on long chain ketones have not tested temperatures lower than 300ºC and it is therefore unclear whether they could form in these situations. These values can be used to propose the existence of a lower heating threshold which is similar to the maximum heating threshold suggested by previous research. Even if it is unclear whether long chain ketones could have been formed without the presence of higher temperatures, it is also possible that their origin was on previous cooking activities performed in the vessels before their involvement in the burial ritual.

Regarding other products, Sample 11, in the Epicardial phase, presented several peaks associated with degradation products from abietic acid (dehydroabietic acid, 6-dehydrodehydroabietic acid, 7-oxodehydroabietic acid, 15-methoxy dehydroabietic acid and retene). The main moiety, methyl dehydroabietic acid, appears after oxidative dehydrogenation (Pollard and Heron 2008) and 7-oxo dehydroabietic acid and retene are commonly found in aged and heated coniferous resins. Nevertheless, small quantities of these molecules have been reported in many archaeological residues, thus suggesting resins could have been a product commonly contained in pottery or they could have been unintentionally incorporated into the vessel potentially from woodsmoke during use (Oras *et al.* 2017, Reber *et al.* 2018) or by soil contamination. Additionally, sample CS11 presents a wider range of diterpenoids such as isoprimaric acid and molecules which could have originated from further oxidation (15-methoxy dehydroabietic acid) (Courel 2016) and dehydrogenation (methyl 6-dehydrodehydroabietate) (Colombini *et al.* 2005). This would suggest that a more complete molecular fingerprint indicating the presence of degraded coniferous resins possibly as a residue originating from vessel use should not be rejected (Figure50). In this case, diterpenoids are accompanied by a range of free fatty acids, cholesterol and an unknown cholesterol derivative, which could suggest the presence of a mixture of various products resulting from the participation of the vessel in diverse tasks.

These range of products (heated animal fats and a possible coniferous resin residue) would present certain continuities across the sequence in Can Sadurní with mainly the presence of degraded animal fats, sometimes heated (CS5) (Table 33). Moreover, sample 14 in layer 10b (NP2) and sample 20 in layer 11a3 (NP1b) presented a significant molecular profile which had also been found by Spiteri (2012) in the Early Neolithic layers. Mainly fatty alcohols (C24 to C27 in CS14 and C24 to C29 in CS20) and very long chain fatty acids (C24 to C30) were accompanied by a series of hydroxy fatty acids with 15-hydroxy

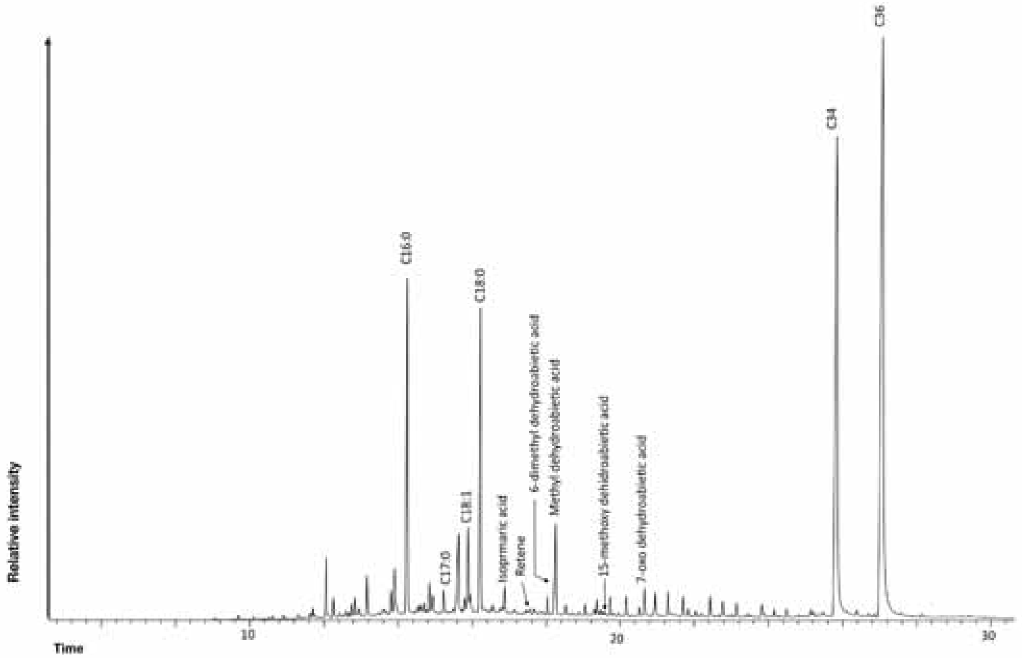


Figure 50: Chromatogram from sample CS11 (Epicardial) presenting a molecular fingerprint characteristic of conifer resins. C34: Tetratriacontane (internal standard), C36: Hexatriacontane (internal standard).

hexadecanoic acid as the main moiety. Others were 17-hydroxy octadecanoic acid, 19-hydroxy eicosanoic acid, 21-hydroxy docosanoic acid, 23-hydroxy tetracosanoic acid, 25-hydroxy hexacosanoic acid, 14-hydroxy hexadecenoic acid, 16-hydroxy octadecanoic acid and 18-hydroxy eicosanoic acid. Moreover, four extra peaks presenting a characteristic 117 m/z ion for ω-1 hydroxy groups and a 149 m/s ion usually attributed to diols were temptativelly identified as 1,23 tetracosanediol, 1,25 hexacosanediol, 1,27 octacosanediol and 1,29 triacontanediol (Figure 51). Given that traces of wax esters were identified in sample 20, it should not be ruled out that the identified alcohols, diols and hydroxy fatty acids resulted from the hydrolysis of a wax. The resulting products would not be incoherent with the structure of beeswax mono, di and hydroxyesters (Tulloch 1973) The presence of 15-hydroxy hexadecanoic acid is coherent with the structure of hydroxyesters from beeswax (Tulloch 1971) and has been previously reported in hydrolised beeswax archaeological samples (ex: Copley *et al.* 2005d, Correa-Ascencio and Evershed 2014). Nevertheless, the possibility that similar patterns could be expected from plant waxes should not be fully rejected. Furthermore, the absence of any significant quantities of alkanes suggests further DCM/MeOH might be necessary to evaluate whether the residue could have originated from a product different from degraded beeswax.

To further characterise possible animal fats detected in the Early Neolithic layers, samples CS9, CS10 and CS12 were submitted to compound specific isotopic analyses.

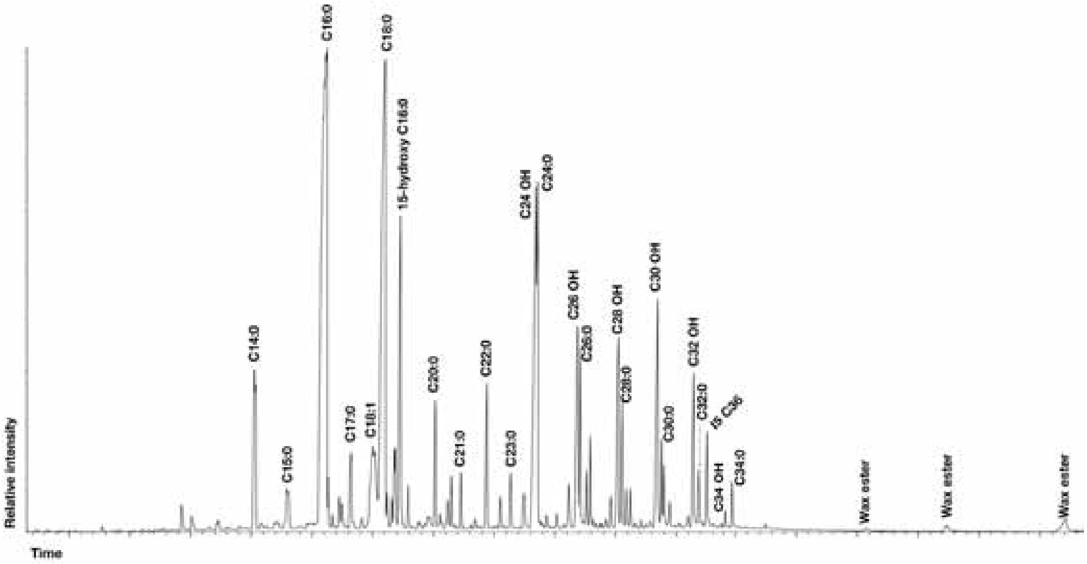


Figure 51: Chromatogram from sample CS20 (Postcardial). Presents a fingerprint characteristic of a hydrolysed wax with some peaks attributed to complete wax esters. It is important to notice the intenisty of the peak attributed to 15-hydroxy hexadecanoic acid.

Results seem to present values similar with previously obtained data from Spiteri *et al.* (2016). In this case, CS12 would present a $\Delta^{13}C$ value of 0.1, which would highly suggest the presence of major amounts of non-ruminant adipose fats should not be rejected. Alternatively, CS10 seemed to be coherent with values attributed to dairy products and CS9's results would suggest the presence of significant quantities of ruminant adipose fats in this vessel (Figure 52).

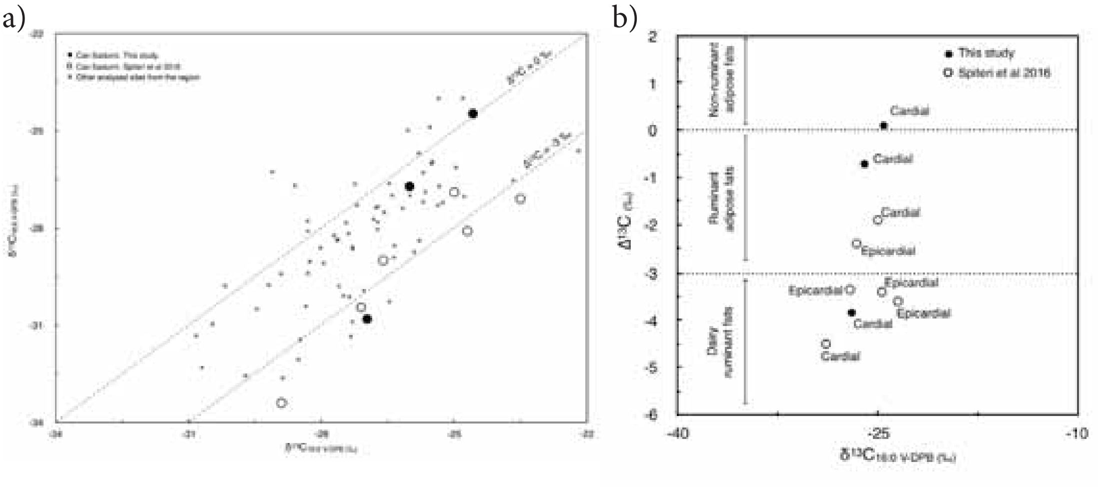


Figure 52: Results from the compound specific isotopic studies. a) plot presenting the $\delta^{13}C$:18:0 and $\delta^{13}C$:16:0 values b) plot presenting the $\Delta^{13}C$ and $\delta^{13}C$:16:0 values.

Given that it can be assumed that residues would result from the accumulation of lipids after repeated uses, a Bayesian mixing model incorporating dairy, ruminant and non-ruminant end members was used to approximate the importance each product could have had in Can Sadurní. Initially samples from this study are taken into account and they are later compared with the overall results from Spiteri *et al.* 2016.

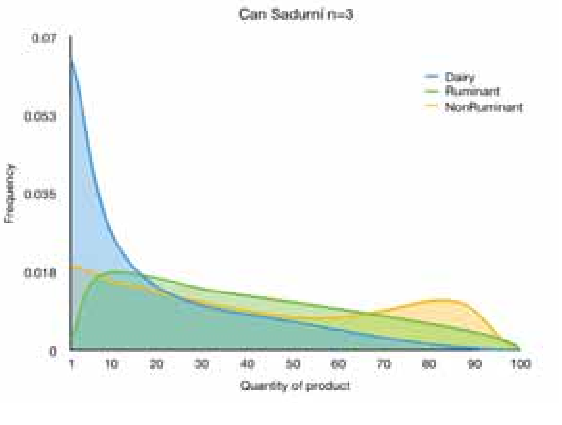


Figure 53: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all samples where compound specific $\delta^{13}C$ isotopic ratios were measured.

For the thee samples analysed in this study (Figure 53), it would seem that non-ruminant adipose fats could have played an important role (median=36%). Alternatively to what might be suggested by the absolute δ¹³C and the Δ¹³C values, the model does not seem to attribute any major weight to dairy products (median=12%) but considers ruminant adipose fats (median=35%) to be significantly present. Given the small sample size and the fact that both samples CS12 and CS9 plot fairy close to the non-ruminant area, the model's suggestion of a high importance of non-ruminant animal fats is to be expected. Nevertheless, the relevance of the existence of dairy products in Can Sadurní must not be understated. When incorporating data from Spiteri *et al.* (2106), the percentages of each product product significantly vary.

When all available data including samples published by Spiteri *et al.* (2016) are taken into account (Figure 54), the model significantly increases the importance dairy products might have had. Although ruminant adipose would be the product with the highest probability of being found in vessels with no mixtures, dairy is the source with the second highest median (28%), followed by non-ruminant adipose fats (median=23%). Ruminant adipose fats would be present in significant amounts as well (median= 35%).

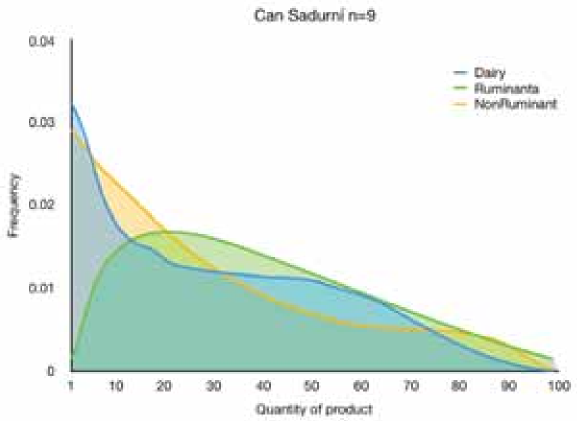


Figure 54: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all available samples from Can Sadurní, including data fron Spiteri *et al.* 2016.

In conclusion, the most complete model does not support rejecting the presence of major quantities of dairy ruminant fats in the vessels but does not rule out the importance of non-ruminant products in some of the vessels.

5.2.6 Conclusions

Organic residue analyses from samples across the stratigraphic sequence at Can Sadurní reveal that ceramic vessels from both the Early Neolithic and later periods had been used for a range of tasks including storing and heating animal fats and containing resins and waxes (possibly beeswax). Further isotopic analyses on selected residues by Spiteri and this study reveal that a significant amount of vessels could have included residues related with the processing of dairy products. In the case of CS10, recovered in layer 13, the detection of ω-(o-alkylphenyl) octadecanoic acids could suggest that the pot possibly had multiple uses, some involved in the processing of raw milk to produce dairy products and other related with cooking at high temperatures. Alternatively, were these two markers interpreted as originating from the same use, it is also possible that dairy products were included as an ingredient of the everyday recipes from that time period.

Regarding animal fats, their presence is coherent with the use-wear patterns in lithic tools, which also provide data on the labour involved in processing meat products. The presence of more adipose fats plotting at the ruminant area than the non-ruminant area is expectable in Can Sadurní given the low percentage of swines amongst bone remains. Nevertheless, when a valid non-ruminant residue has been detected (CS12) its $\delta^{13}C$ isotopic values are coherent with data published by Navarrete *et al.* (2017). Studies on bone collagen suggested these species would have been home-base managed, possibly ingesting leftover food from the human diet, which would not be incoherent with the detected isotopic values in the residues. Given this results so far it is not possible to evaluate possible herd exchanges where animals raised following a certain management strategy would be consumed by a group applying different foddering practices. Nevertheless, in the future, a comparison of said differences between bone collagen on pigs from the an archaeological site and residue lipids in pottery from the same context might be a powerful tool to explore the possible importance of herd exchange in this period.

Regarding plant residues, the extent of its importance remains difficult to assess the extent of its importance given the difficulty in detecting significant molecular markers to infer its presence. Although it should not be rejected that, following Spiteri (2012), those samples with minimal amounts of residues could have contained plant material, the absence of specific biomarkers and the fact that animal residues can easily mask their signal given their higher intrinsic amounts of fats, makes their study more difficult. Nevertheless, in the present study, sample CS11 yielded a signal which can be associated with the presence of resins (possibly from a coniferous species). This product would have been one of the main sources of adhesives in prehistory. Resins have been usually identified as residues in the hafting areas in lithic tools in the Mediterranean region (Regert *et al.* 2000) and, therefore, any Early Neolithic group would have needed a certain amount of it for everyday use repairing and producing the tools needed to perform the myriad of activities related with hunting, harvesting, farming and herding. In consequence, the possibility that the vessel was used as a collector of resin from a bleeding tree should not be ruled out. Moreover, the existence of an applied appliquée close to the rim of this vessel could have facilitated its attachment to a tree. Alternatively the use of the vessel to produce tars must be also taken into account (Pollard and Heron 2008) although no triterpenoids were recovered from the analysed vessel.

In conclusion, the range of products associated with the studied ceramic vessels in Can Sadurní indicates pottery was being actively used to transform organic material originating in the immediate environment. Absence of biomarkers for marine residues might be expectable when the situation of the cave in the middle of the Garraf Massif is considered. Even after taking into account that the sea would have been significantly closer due to the absence of the Llobregat delta, the cost of ascending and descending the mountain might have acted as a deterrent for the easy arrival of said products. Although access to marine seashells and their use as ornaments indicates this ecological niche was not being ignored, had seafood ben consumed in Can Sadurní either their preparation could have avoided the use of pottery or their low frequency prevented the formation of any meaningful and detectable residues.

5.3 La Serreta

5.3.1 Introduction

The La Serreta is an open air site near Vilafranca del Penedès, 205 meters above the sea level and approximately 20km from the coastline. The site was excavated under contract work during the construction of tolls at several junctions in the AP7 highway. In 2008 a surface survey detected several areas of archaeological interest and, in 2009, several prospection trenches were dug to detect and evaluate the extent of possible archaeological sites. As a result, several prehistoric and historic features were detected. The excavations at the La Serreta site, corresponding with the Vilafranca centre junction, revealed 89 negative structures 4 of which belonged to the Early Neolithic (Esteve *et al.* 2010). These were silos, found close together, presenting a good preservation state given that the closing curve was still present in one of them. The contents of said features were highly variable, ranging from remains of domestic debris to layers of gradual elluviation.

5.3.2 Archaeological features relevant in this study

E59 was a semi-oval feature 110cm in maximum diameter and 15cm of depth with two layers of heat-altered and fractured stones. Apart from pottery, a core fragment, retouched flint blades and macrolithic fragments were recovered from there. No animal bones were present in this feature. Charcoal originating from an angiosperm organism dated from 6410±40 BP. At least 15 vases from 18 pottery fragments still preserved handles, plain appliqués and two sherds with cardial decoration (Esteve *et al.* 2010).

E61 had a globular shape and a truncated cone section with a maximum diameter of 85cm in the top and a depth between 35 and 45 cm. A range of 6 deposition layers were composed by stone blocks similar to feature E59. Moreover, two flint cores and blades from jasper and flint were only found apart from pottery (Esteve *et al.* 2010). Charcoal originating from Arbutus unedo, which can be considered a short lived sample, dated from 6490±40 BP (Oms *et al.* 2014). 31 pottery fragments from at least 15 vases featured a significant amount of cardial decorated pottery both on appliqués and on the wall (Oms 2014).

E79, measuring 135cm of maximum diameter and 30cm of depth, contained 3 layers composed of fractured and heat-altered stones. Fragments of grinding stones and a hyaline quartz blade were the non-ceramic materials recovered in this feature (Esteve *et al.* 2010). Charcoal originating from Arbutus unedo, which can be considered a short lived sample, dated from 6420±40 BP (Oms *et al.* 2014). 23 fragments from at least 21 vases featured plain appliqués and two sherds with cardial decoration (Oms 2014).

E75 also presented a truncated cone shape deduced from the feature's good preservation state. Pottery from the refillings was associated with the Epicardial period and featured impressed decoration and grooved features on appliqués or on the wall, sometimes combined with decoration performed with a piercer (Esteve *et al.* 2010). Charcoal originating from Quercus sp. Dated from 6160±40BP (Oms *et al.* 2014). No samples for residue analysis were taken from this feature.

5.3.3 Archaeological materials

5.3.3.1 Pottery remains

The study of the ceramic assemblage performed by Oms (2014) indicates the presence of middle or big sized mainly ovoid or spheric vessels accompanied by a hemispheric/cylindric pots and two necked vases. Related to the production practices, surface colours are coherent with irregular firing and the clay contained calcium carbonate inclusions with also some mica and quartz. The vessel surfaces were either polished, smoothed or, in one case, spatulated. Cardial decorations either on the wall or on appliqués featured up to eight simple and two complex motives. As suggested by previous research (Oms *et al.* 2016a) the geographic similarity makes La Serreta a site possibly related with Guixeres de Vilobí and Cova de la Guineu. Both sites have also been included in this study.

5.3.3.2 Animal and plant remains

As indicated, no animal bones could be recovered from the studied features and plant remains beyond the charcoal used for dating were significantly scarce and hitherto not discussed in the consulted literature.

5.3.4 Sampling and analytical techniques

Obtaining data regarding the possible pottery contents in the site of La Serreta offers great potential to acquire insights into vessel use in one of the core Cardial areas across the Catalan seashore. Along with Guixeres de Vilobí, Cova de la Guineu and Cova Sant Llorenç, La Serreta is in a linear transect from the pre-litoral mountain range to the sea. The opportunity to compare pottery use across these sites will help detect possible different economic strategies put in place by the first farmers in the Penedès region. More specifically, given that no animal bones were recovered from La Serreta (Oms, 2014), the isotopic study of animal fats preserved inside the clay matrix may shed some light into the fraction of the husbandry products that were finally processed with a pottery container.

Lab ID	Feature	Layer	Vessel ID	Sherd ID	Shape	Firing	Decoration	Plastic decoration	Weight (mg)
SE1	59	1	1	16	Rim	Oxidised	None	Appliquée	100
SE3E	59	1	3	11	Rim	Mixed	Cardial	None	64
SE5	61	6	5	53	Rim	Mixed	None	Appliquée	98
SE9	61	4	9	10	Wall	Oxidised	Cardial	None	97
SE10	61	3	10	1	Wall	Oxidised	Cardial	None	106
SE12	61	5	12	43	Wall	Oxidised	Cardial	None	105
SE13	61	4	13	32	Wall	Oxidised	Cardial	Appliquée	103
SEP1	61	4	7	31	Rim	Oxidised	Cardial	Handle	107
SEP2	61	6	6	46	Rim	Oxidised	None	Appliquée	100
SEP3	61	6	8	61	Rim	Oxidised	Incised	Appliquée	105
SEP4	61	4	11	27	Wall	Mixed	Cardial	Appliquée	101
SEP5	59	1	4	14	Rim	Mixed	Cardial	None	103

Table 36: Analysed pottery fragments from La Serreta.

To this end, 12 distinct vessels recovered from silos 61 (n=9) and 59 (n=3) were submitted to organic residue analysis (Table 34). While eight samples (66%) presented some type of cardial decoration, six samples also contained either plain, incised or impressed appliqués (50%). Rims were 58% of the assemblage (n=7) and 5 wall pieces (42%) were selected too. Regarding firing, the majority of the selected assemblage was clearly oxidised (66% n=8) while four samples presented more brown to black coloration usually associated with mixed oxygen-rich and oxygen-deprived firing atmospheres. The assemblage presented a range of cardial decorative motives. Mainly featuring horizontal strips with several variations. According to Oms (2014), they were simple (A1), wide (E1), with short descendent strips (D1), with simple downwards meanders (D3) or with descendent oblique strips (H3).

When possible, up to 1g of pottery was drilled. In one case (sample SE3), the desired quantity could not be achieved without compromising the integrity of the pice and only 0.63g could be collected. Sample weights were later taken into account to calculate the total lipid extracts. The resulting ceramic powder was subjected to an acidified methanol extraction and analysed via Gas Chromatography. When retention times were not enough to identify specific molecular species, a selection of samples was analysed by Gas Chromatography - Mass Spectrometry (GC-MS). Finally, those samples meeting the appropriate requirements were submitted to compound-specific isotopic analysis (GC-C-IRMS). All the work was performed at the ICTA laboratories.

5.3.5 Results and discussion

Lab ID	TLE	PS	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
SE1	13.2	2.9	-29.11	-26.27	2.83	Degraded non-ruminant adipose fat
SE3	58.3	0.9	-29.71	-32.56	-2.85	Degraded ruminant adipose fat
SE5	76.7	0.8	-27.37	-30.14	-2.77	Degraded ruminant adipose fat
SE9	9.3	14.9	-	-	-	Degraded possible plant residue
SE10	3.7	4.5	-	-	-	Non-significant
SE12	6.9	10.0	-	-	-	Degraded possible plant residue
SE13	30.9	1.7	-30.83	-31.33	-0.5	Degraded ruminant adipose fat
SEP1	8.1	3.2	-	-	-	Degraded archaeological fat
SEP2	10.8	2.7	-27.95	-29.09	-1.14	Degraded ruminant adipose fat
SEP3	6.7	3.8	-	-	-	Degraded archaeological fat
SEP4	521.2	0.3	-29.46	-30.5	-1.04	Degraded ruminant adipose fat
SEP5	248.8	0.4	-30.18	-29.79	0.39	Degraded non-ruminant adipose fat

Table 37: Results from the lipid extractions and the GC-FID and GC-C-IRMS analyses. TLE: total lipid extract is expressed in µg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio.

Results in La Serreta indicate that 11 out of 12 vessels contained significant quantities of lipids (91%) (Table 35). No statistically significant differences were found between lipid quantities from walls or rims (Mann-Whitney U=14, n=12, p=0.62) and the presence of either appliqués or impressed cardial motives did not seem to affect the amount of residues either (Mann-Whitney U=21, n=14, p=0.74). Nevertheless, a strong correlation was found regarding the sherd coloration (Mann-Whitney U=0, n=12, p=0.008). Samples presenting clearly orange to beige colours were consistently more depleted than brown or generally darker pieces. The box-plot in Figure 55 shows this phenomenon. In this case, it is unclear wether these changes in the surface colour originated from specific firing conditions, the original colour of the clay, or possible events during use such as sooting that would also have affected the sherd.

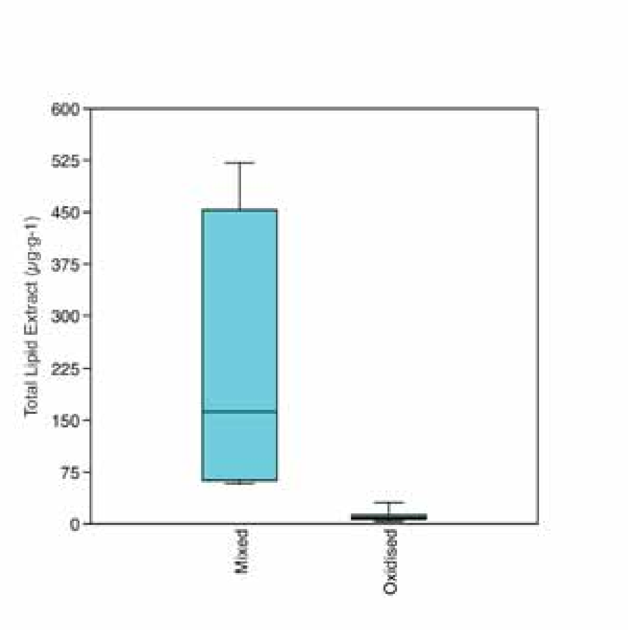


Figure 55: Box plot comparing the total lipid extracts obtained from pottery fired in mixed and in oxidised environments.

The free fatty acid profiles across all samples ranged from C12:0 to C30:0 with the most important molecules being palmitic acid and, in those samples with the highest amount of lipids, stearic acid. Archaeological molecules were accompanied by alkanes and phthalic acids possibly originating from modern contamination and plastic bags. In some cases, (ex: SE3 or SEP4) sherds with high concentrations of lipids contained trace amounts of branched fatty acids, dicarboxylic acids and keto acids. Most of these molecules would have originated from unsaturated fatty acids such as oleic or palmitoleic acids as a result of postdepositional effects such as oxidation and microbial activity (Regert *et al.* 1998).

The ratio of palmitic to stearic acid (P/S) in the case of La Serreta seemed to correlate with the total lipid extract. The samples with the highest yields (SEP4 and SEP5) were also the ones with the lowest P/S ratios and only samples with less than 10µg·g^{-1} (SE9 and SE12) were the ones with up to 15 times more palmitic than stearic acids. These are the highest P/S values encountered across the study. The presence of any significant amount of plant derived lipids can be rejected for at least seven samples (SE1, SE3, SE5, SE13, SEP2, SEP4 and SEP5). This is due to a complete absence of plant biomarkers and a significant amount of stearic acid relative to palmitic acid, which, even after taking into account that C16:0 is slightly more soluble in water than C18:0, would not be coherent with the distribution of fatty acids in plant triacylglycerols. Alternatively, there are no significant evidences against an animal origin. In the case of samples

SE9 and SE12, although specific plant biomarkers are equally absent, significantly low amounts of stearic acid relative to palmitic acid implicate that the possibility they originated from plant triacylglycerols is also possible.

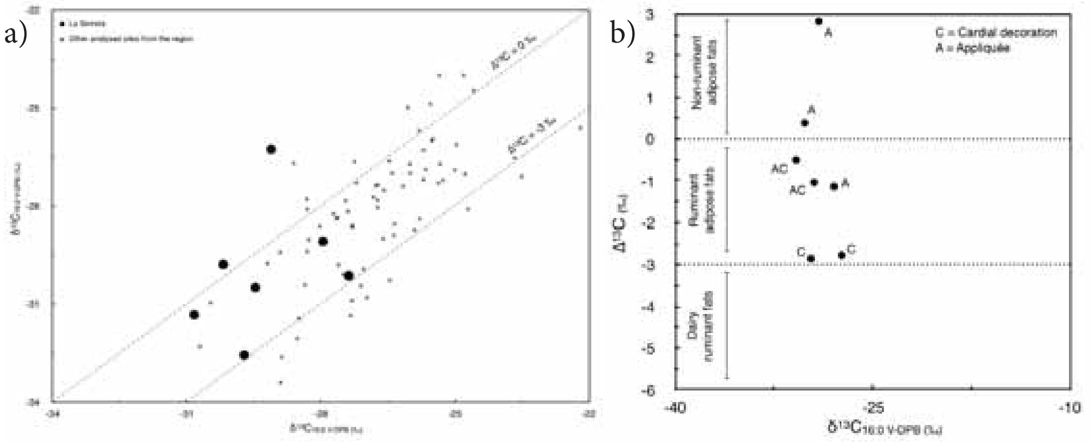


Figure 56: Results from the compound specific isotopic studies. a) plot presenting the δ¹³C:18:0 and δ¹³C:16:0 values b) plot presenting the Δ¹³C and δ¹³C:16:0 values.

After careful evaluation of the P/S ratios, compound-specific isotopic analyses were performed on samples where the presence of plant lipids could be more securely ruled out (P/S < 3). Moreover, there seems to be no correlation between the total lipid extract or the P/S ratio and isotopic values, which would support that all selected sherds may have contained a range of different amounts of degraded animal fats. Following the same patterns observed in both Cova de la Font Major and El Molló, La Serreta presents two samples with a $\Delta^{13}C$‰ close to -3‰, three values plot around -1‰ and two samples (SE1 and SEP5) would be coherent with non-rumiant adipose fats ($\Delta^{13}C>0$‰) although it is not clear wether they could belong to porcine fats (Figure 56).

Initially, there is no evidence to date supporting the presence of dairy products amongst the residues in La Serreta ($\Delta^{13}C < -3$‰). Additionally, although samples SE3 and SE5 present values which could also be coherent with the mixtures of minimal amounts of dairy fats with significant quantities of other animal adipose fats, isotopic values were not enriched enough for marine products to be necessary in order to explain its values. Therefore, were any marine products cooked in the analysed vessels, the amount would have not been significant. While the majority of the samples (71%) plot around an area similar to that of modern ruminant adipose fats, two (29%) were above $\Delta^{13}C = 0$, this isotopic result would not be possible if the residue was composed of any large amount of ruminant fats. Therefore it cannot be rejected that non-ruminant adipose fats where one of the main components from these two values. To clarify this last issue, the two non-ruminant samples from La Serreta have been compared with modern authentic porcine and non-porcine fats. Figure 57 shows that, although the archaeological samples are not significantly different than reference values, they plot at the edges of the porcine fat distribution. A sample of marmot fat from Switzerland presents the same values of SEP5, but this species usually inhabit mountainous areas which makes its presence in La Serreta highly unlikely. The absence of animal remains prevents further interpretation and, therefore, in light of the available data, it seems that there is no evidence to reject the possibility that the recovered non-ruminant fats were significantly composed of porcine adipose fats.

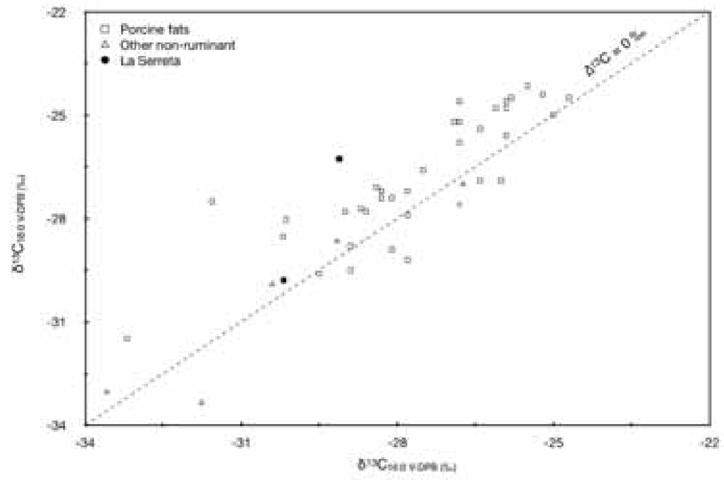


Figure 57: Comparison of reference δ^{13}C16:0‰ and δ^{13}C18:0‰ isotopic values from modern non-ruminant adipose fats with archaeological samples. Reference data obtained from Evershed *et al.* 1997a, Copley *et al.* 2003, Spangenberg *et al.* 2006, Gregg *et al.* 2009, Horiuchi *et al.* 2015, Lucquin *et al.* 2016b, Colonese *et al.* 2017b

In conclusion, the majority of the analysed sherds in La Serreta contained degraded archaeological residues which could be associated with degraded animal fats (58%) and possibly some type of non-animal residue (possibly including plant) (16%). Isotopic analyses indicate that less than 30% of the animal residues would have belonged to non-ruminant species while more than 70% of the studied assemblage was associated with ruminant adipose fats. Assuming all available animal products were processed with pottery, it seems that the sources of meat accessible by the social groups in La Serreta would have been composed by a majority of ruminant and a minority of non-ruminant animals. This is coherent with the composition of bone assemblages from other contemporary Neolithic sites (Nadal *et al.* 1999).

In the case of La Serreta, all the studied samples presented some kind of decoration. Nevertheless, vessels decorated with cardial techniques contained non-ruminant fats (SEP5), ruminant fats (SEP4, SE13 and SE3), residues with high P/S ratios (SE9, SE12 and SEP1) or no residues at all (SE10). Vessels with appliqués were also found to contain non-ruminant fats (SE1), ruminant fats (SE5) and residues with high P/S ratios (SEP3). Different cardial motives did not seem to correlate well with the vessel contents either (Student's T=1.93 n=7 p=0.09). Following these results, it is difficult to support the idea that the cardial decoration had any meaning beyond a pure aesthetic goal. In fact, they did not seem to affect vessel contents at La Serreta. Nevertheless, to fully reject this option a bayesian mixing model has been applied to study possible tendencies in the use of vessels with these two decoration techniques. Given that no clear biomarkers for plant and marine products were present, the end-members used were non ruminant adipose fats and ruminant dairy and adipose fats. It is assumed that the isotopic values of palmitic and stearic acid would be the result of an accumulation of vessel uses. Therefore, had the presence of cardial impressions or appliqués promoted the use of either one of the three considered end-members, it should appear when comparing the accumulated expected probability values for each source.

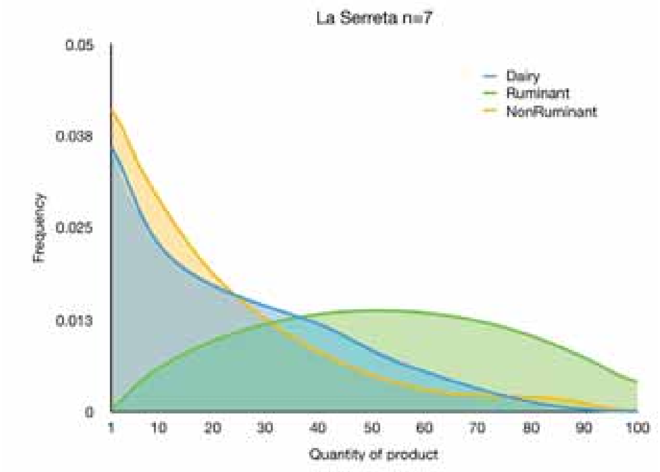


Figure 58: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from La Serreta.

The accumulated results for all the vessels whose compound specific isotopic data was available suggests the presence of mixtures including mainly ruminant adipose products (median=51%) and, in lesser quantities, possibly dairy products (median=22%) and non-ruminant fats (median=16%)(Figure 58). When evaluating only those sherds with an apliqué, the probability distribution does not differ significantly from the overall results. Mainly ruminant adipose fats (median=28%) could have been mixed with other products (dairy=20%, non-ruminant=19%)(figure Xa).

Nevertheless, when selecting only samples with cardial decorations, a significant increase in the presence of ruminant adipose fats (now median=45%) and dairy products (now median=25%) can be noted (figure 59a). Furthermore, the model predicts a minor chance for the presence of high amounts of non-ruminant products for vessels with appliqués (possibly as a result of the high $\Delta^{13}C$ values in two of the samples), which is not present in the Cardial estimates. In fact, the possible presence of non-ruminant fats slightly falls from a median of 19% to a median of 12% between plain appliqués and Cardial decorations. The subtraction of the Plain appliquée values minus the Cardial values provides a means of visualising the differences between the two decorative techniques (figure 60). According to the models, Cardial vessels would present a higher probability of containing smaller amounts of non-ruminant and isotopically enriched ruminant adipose fats. The probability of containing significant quantities of dairy products would be slightly stronger in Cardial vessels. Alternatively, vessels with plain appliqués could have more frequently contained low amounts of dairy products while presenting a higher probability of containing significant quantities of non-rumiant animal fats.

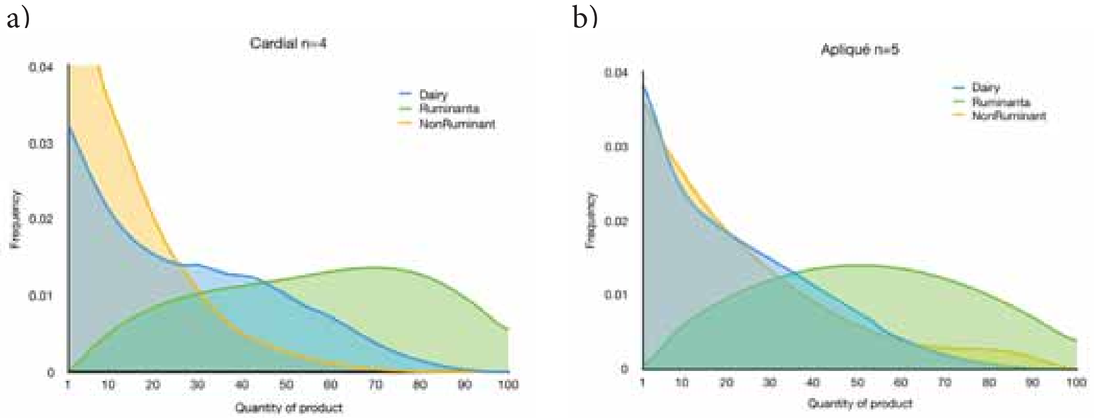


Figure 59: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in a) sample presenting cardial impressions and b) samples presenting appliqués.

Following the model estimates, it would seem that a slight tendency differentiating the use of Cardial imprints and appliqués can not be fully rejected. Nevertheless, these results have to be carefully evaluated. These minor differences seem to appear only on the types of animal fats used. Furthermore, this comparison has been performed with only 4 cases of Cardial wares and 5 cases of plain appliqués. The fact that this is a small sample size implies further residue analyses in La Serreta might sensibly alter these interpretations. So far, a Mann-Withney test between the non-ruminant probability distribution curve provides a result indicating that the null hypothesis that no significant differences exist between cardial and appliqués can be rejected (U=3770, p=0.0026). Nevertheless, changes in the quantities of dairy or ruminant fats do not seem to be statistically rellevant (Dairy: U=4745.5 , p=0.53, Ruminant: U=4761.5 , p=0.56).

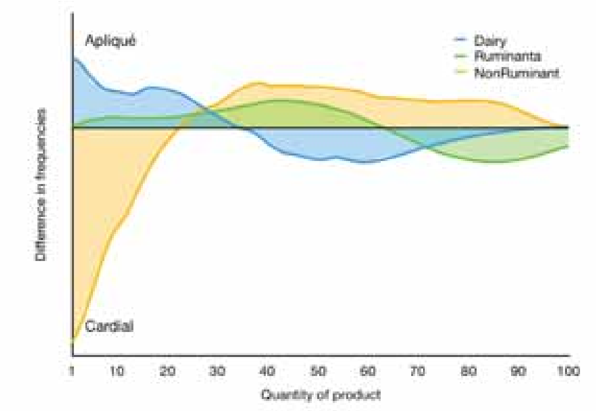


Figure 60: Comparison of modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in samples with Cardial impressions and appliqués.

5.3.6 Conclusion

In conclusion, although the scarcity of other paleoeconomic data in La Serreta poses some challenges for further interpretation of the residues, the recovered data supports the fact that pottery would have been used to contain, at least, a certain amount of degraded animal fats. Specific fatty acid profiles in two samples suggest, despite the absence of clear biomarkers, the possible presence of minor amounts of plant residues should not be completely ruled out and compound specific isotopic analyses reveal the presence of mainly ruminant and non-ruminant adipose fats. Interestingly, although the latter were only present in structure E59, it is difficult to evaluate this possible trend given the small sample size. Finally, after evaluation of the results through a bayesian mixing model, some differences in the nature of the residue could be detected depending on the decoration technique applied in the vessel but further analysis will be needed in order to completely validate its existence.

5.4 Guixeres de Vilobí

5.4.1 Introduction

Guixeres de Vilobí is an open air archaeological site in one of the elevations belonging to the chalky cretaceous outcrops between Vilobí del Penedès and Sant Martí Sarroca. It contained a set of prehistoric structures which were found on a small plateau approximately 100m upslope in an area 10m wide and limited by a small limestone cliff in the north. This prehistoric settlement would have been around 300 meters above sea level and roughly 27km away from the sea.

The site was initially reported by A. Ferrer after detecting archaeological materials during a superficial prospection. Between 1973 and 1974 a systematic prospection and test trenches were performed by Baldellou and Mestres (1981) and the site was extensively excavated during 4 campaigns between 1981 and 1984. After expanding the test trench (cala 1) with the most promising results, a wide excavation uncovered the remains of a significant Early Neolithic site. The archaeological finds and analyses performed in the materials recovered were amongst the first complete studies of a Cardial Early Neolithic habitat and played a major role in shaping the understanding of this historical phase during the 90's (Baldellou and Mestres 1981). New excavations in Guixeres de Vilobí started in 2016 by Josep Mestres and Xavier Oms have been able to expand the stratigraphic and archaeologic knowledge available for this site which is now around 170m^2 in extension (Oms *et al.* in press).

5.4.2 Archaeological features relevant in this study

As reported by Mestres (1981), the site contained a straightforward edaphologic sequence with a superficial layer presenting a dark organic sediment with roots and other bioturbation agents, a second layer with white well-cemented chalky sediment, and a third layer associated with a yellow to white marl bedrock. Further analysis of the artefacts recovered in the site indicated the existence of three occupation phases named horizons A, B and C. Horizon C was dated as a Postcardial or Evolved Early Neolithic, and was associated with a silo 1m in diameter and 80cm deep with no archaeological material inside. Horizon B was limited to a specific space and corresponded with the Epicardial Early Neolithic. Finally, Horizon A contained the beginning of the human occupation in the site during the early Cardial Neolithic. Excavation works from 2015 onwards have found two spaces with ellipsoidal plans surrounded by thermally altered stone blocks which have been tentatively interpreted as possible huts (Figure 61). Combustion structures composed of small charcoal fragments and thermally fractured stones and storage features and postholes of a significant diameter (around 50cm) were both outside and, more importantly, inside these spaces (Oms 2017a). More recently, these two areas have been found to date from two non-synchronous moments in the most early Cardial Neolithic (6655±45 BP for space 1 and 6458±38 BP for space 2, (Oms *et al.* 2014).

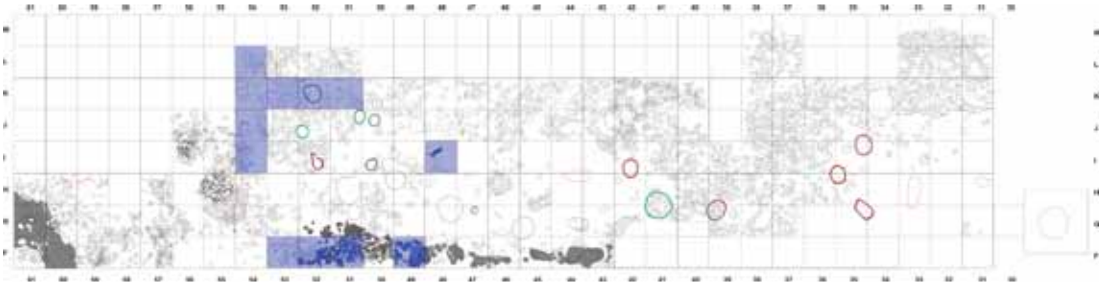


Figure 61: Plan of the excavated area in Guixeres de Vilobí. Blue squares indicate the sampled areas. Source: Oms 2017a.

5.4.3 Archaeological materials

5.4.3.1 Pottery remains

Framed within this archaeologic context, more than 300 pottery sherds had been recovered from the site before new excavations begun in 2015 (Mestres 1981). After analysis by Oms (2014), they could be attributed to at least 85 different vases. This assemblage would have been moulded with clays incorporating limestone, quartz and mica inclusions. The shape repertoire would have included spheric middle sized vessels, necked small vases, big cylindrical vases and hemispheric middle sized vessels. All presented mainly smoothed surfaces with some polished cases and were subjected to a mixed firing technique. Generally speaking, the cardial decoration appeared in spheric and necked vases and in a majority of cylindrical vases. Although the site mainly presented common cardial motives such as the simple horizontal (A1), limited horizontal (A2) and the simple horizontal band with an integrated appliquée (F1) motives, two vases also possibly contained incomplete zoomorphic motives (Oms *et al.* 2016b).

5.4.3.2 Lithic remains

Lithic tools in the site presented a certain technological homogeneity marked by the exhaustive use of raw materials such as flint and jasper. In the case of the pieces attributed to the Cardial Early Neolithic, Mestres (1987) suggested some remains could still present features which could be associated with pre-Neolithic times that would not be present in more recent periods.

Around 2063 lithic remains analysed by Gibaja *et al.* (2018a) suggest that lithic tools, mainly blade supports, were locally produced. This is further supported by the presence of the nucleus' cortex in some pieces. The functional analyses of 182 pieces detected that at least 78 of them had been used. A minimum of 24 showed evidences for the processing of animal matter and three geometric microliths and a notched bladelet sowed impact fractures as a consequence of their use as projectiles. Nine pieces revealed wear related to butchering in both edges, seven were used on dry skin, two on fresh skin and two on bone (Gibaja *et al.* 2018a). Furthermore, nine pieces had been involved in the processing of vegetal matter. Two flakes were related to woodworking, three were associated to cereal harvesting and four could have been used on other non-woody undetermined plants. The tasks performed on the latter involved a variety of movements, which could suggest activities like basketry, rope production and handing implements should not be ruled out. Finally, there was a significant amount of pieces where only the toughness of the worked material could be determined. In these cases, possible uses could include butchering and skinning practices. When the worked matter was found to be tougher and transformed through scraping, plausible interpretations included materials such as wood, bone or antler.

In consequence, the presence of activities that would need to happen across wide ranges of time is not in disagreement with the idea that the settlement in Guixeres de Vilobí was stable across a significant amount of time. A comparison of the data recovered trough use-wear studies across several Early Neolithic sites in the Iberian Peninsula suggests Guixeres de Vilobí fits within a framework of stable and long lived sites with diverse subsistence and artisanal practices on materials from varied natures and food production strategies centred both on agriculture and pastoralism. This is a different situation compared with other sites where hunting and animal herding might have been more important (Gibaja *et al.* 2018a).

5.4.3.3 Animal remains

As a testimony to the range of economic activities performed in the site, a significant amount of animal bones have been recovered since the start of the excavations. The study of the remains found in the 1989 campaign by Josep Maria Miró (1991) pointed out that the assemblage presented a high degree of fragmentation and reported the species of those 118 bones which could be identified for the Cardial and Epicardial periods in the site. These would have been mainly domestic *Ovis aries/Capra hircus* (n=68, 57%), *Sus* sp. (n=11, 9%) and *Bos* sp. (n=10, 8%), but also wild *Oryctolagus cuniculus* (n=23, 19%), *Cervus elaphus* (n=4, 3%), *Bos primigenius* (n=1, 1%), *Mustela* sp. (n=1, 1%), bird remains (n=1, 1%) and fish remains (n=1, 1%). Wild species were interpreted as bing part of a complementary hunting activity.

Anatomically, mainly isolated teeth, followed by metapods, radius and ulnae, were found. Furthermore, specific fragmentation patters found in the site could be linked with the preparation of broths and the extraction of bone marrow (Miró, 1991). These could be "secondary" culinary practices, when cooking with the leftovers of a previous food production process.

5.4.3.4 Plant remains

The complete flotation of all sediment recovered after the 2016 excavation campaign and the studies performed before barely report the presence of any carpological remains. Nevertheless, the presence of several fragments of acorn cotyledon stands out (Buxó 1990).

5.4.4 Sampling and analytical techniques

Taking into account the state of the knowledge about the earliest Neolithic societies in Guixeres de Vilobí, organic residue analysis offers the opportunity to acquire new data on the possible contents of one of the first known ceramic assemblages to date for the Early Neolithic and compare the results with the uses other tools had in order to complete the picture of the different types of labour activities practiced in the site. Furthermore, studies on bone remains will help interpret the types of possible animal products recovered and will facilitate the evaluation of possible differential cooking strategies between species.

To this end, 22 samples from several excavation squares in Guixeres de Vilobí were selected for analysis. These originated from rows L, K, J, I and F and columns 48, 49 and 51-54. Almost all sampled squares could be interpreted as belonging to an outside space immediately adjacent to area 1. Nevertheless, in the case of square I48, collected samples would be placed inside area 1. These were 59% rims (n=13) and 41% walls (n=9) and its colours were consistent with Early Neolithic firing techniques and sampled decorations including cardial impressions (n=2), plain appliqués (n=5) and one incised appliquée. In three cases (GV23, GV58 and GVP5) handles were also present and 54% of the vessels (n=12) were plain (Table 36).

Lab ID	Stratum	Square	Sherd ID	Shape	Firing	Decoration	Plastic decoration	Sample weight (mg)
GV101	IB	L54	101	Wall	Mixed	None	None	100
GV161	II	L54	161	Rim	Oxi	Cardial	None	97
GV20	IB	F51	20	Wall	Mixed	None	appliquée	101
GV23	IB	I54	23	Rim	Oxi	None	Handle	98
GV28F	IB	F49	28	Rim	Mixed	None	None	100
GV28I	IB	I54	28	Rim	Oxi	Cardial	appliquée	102
GV28K	II	K54	28	Rim	Oxi	None	None	83
GV36	IB	F49	36	Rim	Mixed	None	None	107
GV42	IB	F52	42	Wall	Mixed	None	appliquée	104
GV43	IB	L54	43	Rim	Mixed	None	appliquée	100
GV44	IA	J54	44	Rim	Oxi	None	None	99
GV48	IB	F51	48	Rim	Mixed	None	None	82
GV54	II	L52	54	Rim	Mixed	None	appliquée	101
GV58	IB	L53	58	Wall	Oxi	None	Handle	100
GV68	II	J54	68	Rim	Mixed	None	None	102
GV78	IB	L51	78	Wall	Oxi	None	None	101
GV98	IB	F53	88	Wall	Mixed	None	None	110
GVP1	IA	I48	Sieve	Wall	Mixed	None	None	102
GVP2	IB	F52	Sieve	Wall	Mixed	None	None	97
GVP3	IB	F52	Sieve	Wall	Mixed	None	None	99
GVP4	IB	L54	39	Rim	Oxi	Incision	appliquée	105
GVP5	IB	I54	29	Rim	Mixed	None	Handle	108

Table 38: Analysed pottery fragments from Guixeres de Vilobí.

Organic residue analyses were performed on roughly 1g of pottery. Lipids were extracted from samples following an acidified methanol extraction and separated under Gas Chromatography. When retention times were not indicative enough of the molecule's nature, selected cases were analysed by Gas Chromatography - Mass Spectrometry. Finally, compound specific isotopic analyses (GC-C-IRMS) were performed on some of the vessels. All analyses were carried out in the ICTA laboratories.

5.4.5 Results and discussion

It seems that, although the most aggressive extraction techniques were implemented, the degree of lipid matter preservation in Gixeres de Vilobí was amongst the lowest in the study (Table 37). Around 55% of the analysed vessels (n=12) contained lipid amounts lower than the 5μg·g^{-1} threshold for significant archaeological residues. This meant that only 10 samples (45%) could be considered to contain quantities which could be interpreted as originating from the lipid products the vessels could have contained. Nevertheless, samples GV44 and GV23, presented two of the highest amounts of lipids detected so far for the cardial Early Neolithic (GV44 = 893.6μg·g^{-1} and GV23 = 1587.3μg·g^{-1}) and around 100 times more lipids than the remaining archaeologically significant residues, thus suggesting in some cases, fatty materials had been extensively used in the site. Statistical tests suggest (see Table 38) that the hypothesis that vessels with different shapes or different decoration contained different amounts of fat can be rejected. Nevertheless, it is possible that a weak association existed with the piece's original edaphological stratum (see table x). It would seem that samples in layer II did not yield more than 8μg·g^{-1}, layer IB contained a whole range of concentrations from the lowest to the highest amounts and, finally, the two samples from layer IA were amongst the highest. It is not yet clear why samples from layer II should be more depleted in lipids, nevertheless it is possible that these weak association was an artefact of the small quantity of samples taken from layers IA (n=2) and II (n=4), or maybe distinct post-depositional processes affected lipid preservation, or (the less likely option) pottery used in stratum II were less exposed to fatty materials than pottery in stratum 1A.

Regarding decorations, the two analysed sherds with cardial impressions in Guixeres contained non significant amounts of fat and only half (n=3) the vessels with appliqués and half the samples from plain sherds (n=6) contained interpretable lipids. Furthermore, the presence of handles did not seem to affect the amount of residues: sample GVP5 had one of the lowest lipid yields, sample GC23 had the highest lipid yield and sample GV98 contained only 0.45μg·g^{-1} less that the median of the assemblage. In consequence, it seems that the ceramic assemblage in Guixeres de Vilobí was indistinctly involved in a range of uses which might or might not have involved fatty materials. Moreover, it does not seem that the sample points in the vessel or the decoration are good indicators for the presence or absence of significant decoration are good

LAB ID	TLE	P/S	$\delta^{13}C16{:}0$‰	$\delta^{13}C18{:}0$‰	$\Delta^{13}C$‰	Interpretation
GV101	1.7	1.2	-	-	-	Non significant residue
GV161	3.8	1.1	-	-	-	Non significant residue
GV20	2.6	1.4	-	-	-	Non significant residue
GV23	1587.3	1.1	-	-	-	Degraded animal fat
GV28F	11.7	0.9	-30.7	-32.3	-1.58	Degraded ruminant adipose fat
GV28I	1.8	1.5	-	-	-	Non significant residue
GV28K	2.7	0.9	-	-	-	Non significant residue
GV36	1.6	1.6	-	-	-	Non significant residue
GV42	5.2	0.9	-28.35	-30.42	-2.07	Degraded ruminant adipose fat
GV43	9.8	1.3	-	-	-	Degraded animal fat
GV44	893.6	0.6	-	-	-	Degraded animal fat
GV48	2.4	0.9	-	-	-	Non significant residue
GV54	1.3	1.1	-	-	-	Non significant residue
GV58	3.1	1.2	-	-	-	Non significant residue
GV68	6.6	1.2	-	-	-	Degraded animal fat
GV78	1.7	1.6	-	-	-	Non significant residue
GV98	3.0	1.5	-	-	-	Non significant residue
GVP1	12.5	0.7	-28.48	-31.44	-2.96	Degraded ruminant adipose fat
GVP2	12.9	0.9	-28.87	-32.63	-3.76	Degraded ruminant dairy fat
GVP3	6.9	1.0	-	-	-	Degraded animal fat
GVP4	10.5	1.4	-	-	-	Degraded animal fat
GVP5	1.4	1.2	-	-	-	Non significant residue

Table 39: Results from the lipid extractions and the GC-FID and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

indicators for the presence or absence of significant amounts of residues. Had there been an active criteria to use certain vases for lipid-containing products and other vessels for non-fatty materials, it does not seem to be apparent in the analysed samples.

Statistical tests	TLE	P/S
Strata	K-W p(same) 0.12	ANOVA p(same) 0.06
Sherd shape	M-W p(same) 0.94	T test p(same) 0.89
Decoration presence	M-W p(same) 0.66	T test p(same) 0.26

Table 40: Statistical tests evaluating the possible effects of strata, sherd shape and the presence of decoration on total lipid extracts and P/S ratios.

Finally, when taking into account that samples from spatially and temporarily similar archaeological sites have achieved higher preservation rates (ex: Cova de la Guineu, La Serreta), it should not be rejected that specific post-depositional effects from the chalk rich underground environment in Guixeres de Vilobí might be a major factor driving this phenomenon. Nevertheless, it would be also plausible that, similarly to the nearby wide range of uses for lithic tools in south-west of area 1, pots could have been used for a variety of activities which might or might not have involved fat rich products.

Regarding the nature of the residues themselves, all samples presented a range of saturated free fatty acids having from 10 to 26 carbon atoms in its chain. Alkanes and phthalate plasticisers were minor constituents indicating a certain amount of modern contamination originating from plastic bags. Oxidation products such as dicarboxylic acids and keto acids were detected in 60% of the positive samples (GVP1, GVP2, GV23, GV28F, GV43 and GV44). These molecules would point at the cleavage of the double bond in unsaturated fatty acids, thus certifying these types of molecules had been degraded. All samples contained minor quantities of oleic and palmitoleic acids and major similar quantities of palmitic acid and stearic acid. The ratio between these last two chemical compounds (P/S) was never lower than 0.6 and higher than 1.6, with these values, the possibility that the detected range of free fatty acids were coherent with the hydrolysis and degradation of animal tryacylglicerols can not be ruled out.

In the case of Guixeres de Vilobí and contrary to what has been detected in other nearby contemporary archaeological sites (ex: La Serreta and Cova de la Guineu), there are no sherds with two or more times the amount of palmitic than stearic acid. The absence of this phenomenon, which is present (but not exclusively) in degraded plant residues (Copley *et al.* 2005e, Dunne *et al.* 2016), suggests the hypothesis that the analysed vessels in this site were exposed to roughly the same types of products must not be discarded. Although there is a slight tendency favouring higher relative amounts of palmitic acid when overall lipid yields are lower, the r^2 value from the resulting regression line (linear model built excluding samples GV44 and GV23 because they contain around 100 times more lipids than the remaining positive results) indicates that this correlation can only explain less than 30% of the overall P/S variation (see figure 62).

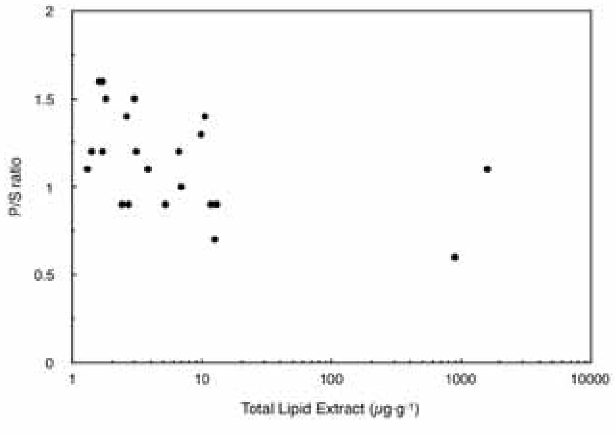


Figure 62: Variation of P/S ratios as afunction of total lipid extracts

Therefore, were there any possible postdepositional effects affecting both the total amount of recovered fats and the relative quantities of palmitic and stearic acids, it could be considered that they did not significantly affect the recovered residues. Moreover, statistical tests suggest not rejecting the null hypothesis that archaeological layers, sherd shape and the presence of decoration are not correlated with the relative amounts of palmitic and stearic acids.

Minor molecules found in some of the samples might provide indices to further specify the type of products contained by the studied pots. Phytanic acid is the result of the bacterial oxidation and hydrogenation of phytol, an important constituent of chlorophyll. It has been used by several researchers to point at the possibility that residues might originate from ruminant fats (Lucquin *et al.* 2016a) although it is also possible that bacteria attacking plant phytol after the sherd had been buried could have also generated this molecule. In Guixeres de Vilobí, chromatograms from samples GVP1 and GVP2 contained peaks whose mass spectra matched the one from phytanic acid (Figure 63).

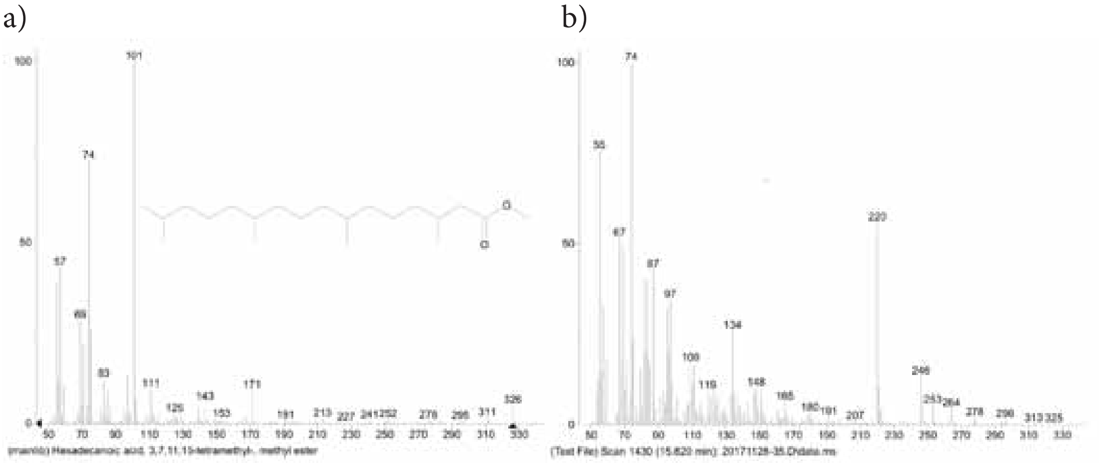


Figure 63: a) reference mass spectra of pytanic acid b) mass spectra obtained at Rt= 15.82 min from sample GVP2.

Branched chain fatty acids have also been used as a possible marker supporting the presence of ruminant fats (Evershed 1993, Mottram *et al.* 1999). The amount of microbial activity in the gut has been associated with the quantity of branched chain fatty acids available. Given the significantly higher amount of bacterial breakdown processes taking place in the ruminant gut compared with non-ruminants, it is expectable that the relative amount of the isomers of C17:0br fatty acid in relation to stearic acid could be a good indicator of this phenomenon (Dudd *et al.* 1999, Hjulström *et al.* 2008). Nevertheless, it has to be taken into account that, following Dudd *et al.* (1999), the consumption of whey or other products containing higher amounts of C17:0br fatty acids by non ruminant animals would also generate the same signal, as branched fatty acids would have been unaffectedly carried over from the diet. The same authors also point out that an exogenous bacterial source should not be ruled out. In the case of Guixeres de Vilobí, the relative amounts of branched chained C17:0 fatty acids varied across the samples set. GVP1, GVP2 and GV23 contained values higher than 0.03, but further data is needed in order to reject or accept the hypothesis that these vessels contained mainly fats originated in a ruminant animal.

To better characterise the recovered residues, compound specific isotopic analyses were performed on a subset of samples meeting the appropriate requirements (GVP1, GVP2, GV28F and GV42). Results indicate that 75% were coherent with ruminant adipose fats (GVP1, GV28F and GV42) while one sample (GVP2) presenting a $\Delta^{13}C$ value lower than -3‰ most probably contained a dairy product. This suggests that animal husbandry in Guixeres de Vilobí was not only focused in the acquisition of meat (which has been attested by three residues containing ruminant adipose fats, use-wear analysis of lithic tools and the study of the animal bone assemblage), but also in milking some of the ruminant animals and incorporating this source of protein to the diet. The genetic modification allowing for lactase persistence and, therefore, the ability to easily digest fresh milk, hasn't been hitherto detected in Early Neolithic individuals from the Iberian Peninsula (Olalde *et al.* 2018 and 2019). In consequence, it is highly unlikely that milk was consumed without any previous treatment which significantly reduced the amount of lactose. To separate the curd from the whey, milk is usually gently warmed before adding an acidifying agent and pottery vessels would be a perfect fit for this step. Taking these evidences into account, it is not possible to discard the possibility that the vessel analysed as sample CVP2 was used in the process of preparing dairy products.

Compared with the rest of the vessels from other archaeological sites in this study, the $\delta^{13}C$‰ absolute values in Guixeres de Vilobí seem to be amongst the most depleted (Figure 64). Given that isotopic values are strongly affected by the organism's diet, it should not be ruled out that animal fats transformed and consumed in Guixeres de Vilobí could be associated with a specific set of environmental conditions affecting the ruminant's fodder. If assuming this as the major factor for isotopic variation, it could be speculated that all the recovered fats would be coherent with a range of animals following a similar feeding pattern, maybe concentrated in a specific area close to Neolithic settlement.

Nevertheless, to fully evaluate this hypothesis, further data on feeding practices should be acquired through collagen δ¹³C and δ¹⁵N isotopic studies and micro-dental wear analyses, amongst others.

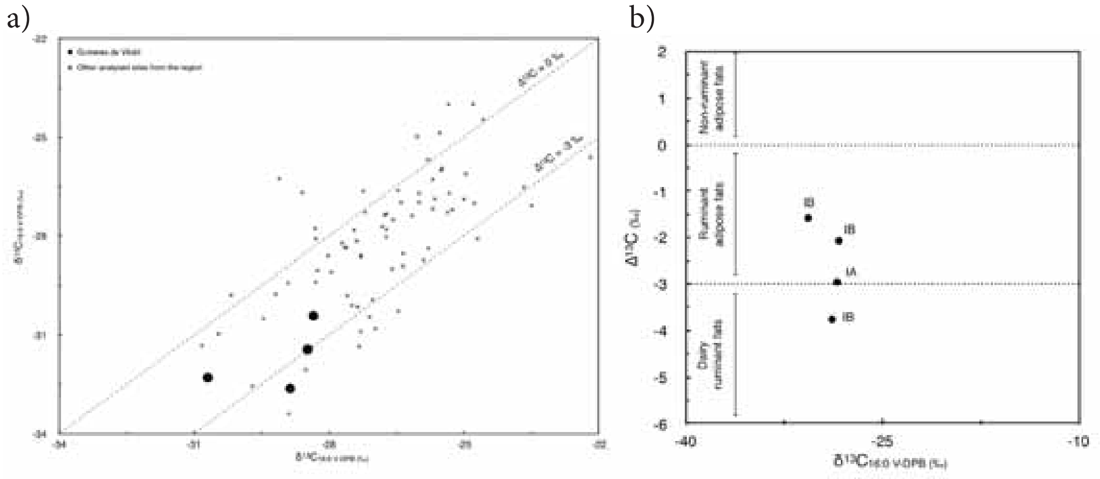


Figure 64: Results from the compound specific isotopic studies. a) plot presenting the δ¹³C:18:0 and δ¹³C:16:0 values b) plot presenting the Δ¹³C and δ¹³C:16:0 values. Labels indicate the layer where the samples were located.

Looking at the available data for the composition of the herd in Guixeres de Vilobí, ruminants were 70% of the assemblage in the site (n=83) and non-ruminants just reached 10% (n=12) (Miró 1991). Moreover, sheeps and goats were the majority species in the ruminant group, which suggests the dairy signal found in samples CVP2 could have originated from one of these species. Given that compound specific isotopic analyses could only be performed on four residues, it is not possible to discard the hypothesis that the results are coherent with the faunal assemblage in the site. Had non-rumimants been cooked in pottery and not exclusively spit roasted, further analyses might reveal evidences for non-ruminant products.

To evaluate the possible existence of non-ruminant isotopic signals masked by ruminant and dairy fats, a bayesian mixing model has been applied to the available data. Plant and marine end members have not been incorporated due to the absence of specific biomarkers for these products. Alternatively, dairy, ruminant and non-ruminant have been considered. The model assumes the isotopic values of palmitic and stearic acid result from an average of the repetitive use of known products and estimates which range of mixtures could have produced the obtained value. The probabilities for each vessel can then be added and normalised to approximate the importance each product could have had in the studied assemblage. Nevertheless, in the case of Guixeres de Vilobí, the small sample size needs to be taken into account as possible future studies might significantly alter the results and interpretations obtained hitherto.

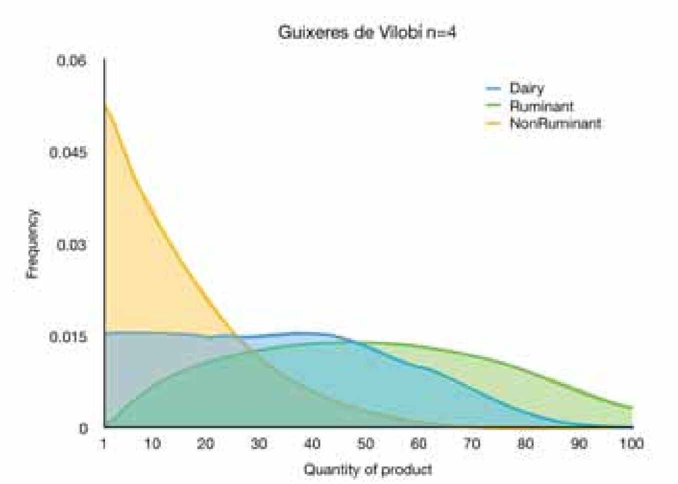


Figure 65: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all samples from Guixeres de Vilobí.

Modelled results from Guixeres de Vilobí are shown to most probably contain lower quantities of non-ruminant (Figure 65). Conversely, a mixture of ruminant adipose fats and dairy products would appear to be the most common and feasible solution. This situation is also observed when the medians for each distribution are taken into account. Non-ruminant (12%) present values significantly lower than ruminant dairy (34%) and ruminant carcass fats (37%).

5.4.6 Conclusion

To conclude, Guixeres de Vilobí presents two samples with lipid quantities amongst the highest across all pottery from this study. Nevertheless, it is also the site which so far presented the lowest residue success rate in this study. The fact that 45% of the samples still presented traces of degraded animal fats indicates that preservation might not be an issue and that it is not impossible that practices involving fat-rich animal products were present in the site. Although all samples with significant amounts of lipids were coherent with animal fats, this pattern seems to differ from other Early Neolithic sites, where higher P/S ratios could imply a minimal input of plant residues can not be fully rejected. Finally, according to molecular and isotopic data, it seems impossible to reject that the majority of the recovered residues would have been composed of a significant amount of ruminant animal fat and, in one case, a dairy product. Taking in to account the sample size, it is not impossible these results were coherent with the bone assemblage in the site.

When looking at the spatial location of all the samples, the area defined by row K and column 54 contained 14 studied pottery sherds while 8 analyses were performed from remains found the area defined by row F. Although both groups present similar preservation rates and P/S ratios, exceptionally lipid enriched samples were detected in adjacent squares (I54 and J54). This result suggest the existence of a possible spatial pattern affecting certain pottery uses which would have involved a significant quantity of lipids. Nevertheless, the study of possible trends which may reflect the use of the space will need further work increasing the amount of positive samples and the studied surface.

5.5 Cova de la Guineu

5.5.1 Introduction

The Guineu Cave is in Font-Rubí, a town in the pre-litoral mountain chain. It is 725 meters above the sea level and 35km away from the sea. The cave is an overture which resulted from the dissolution of cracks in the limestone (Bergadà 1998a) due to karstic processes and its geological surroundings contain red sandstone, limestone and dolomites. It was initially excavated in the 70's by amateurs from the "Associació d'Estudis Científics i Culturals de Mediona" until 1981, when governmental intervention stopped the activity. In 1983, the Museum of Vilafranca resumed the archaeological work and, in 1988, the SERP research group from the University of Barcelona joined the research team (Bartrolí *et al.* 1992). Since then, fieldwork has constantly taken place every year in the cave. Moreover, in recent years, modern archaeological excavation campaigns have focused on the exterior of the cave to fully deplete the stratigraphy (Equip Guineu 1994, Morales *et al.* 2013).

5.5.2 Archaeological features relevant in this study

The cave presents two different depositional dynamics. The upper stratigraphy contains slope deposits and the lower stratigraphy was formed by a karstic dynamic system with the presence of dissolution clays and colluvium deposits (Berguedà 1998).

The most ancient occupations in Cova de la Guineu were sporadic but intense hunter-gatherer visits across the Palaeolithic. In posterior times, the cave was occupied again during the Early and the Postcardial Neolithic (5500-4000 cal BC)(layer IIC) and, starting in the fourth millennia, it would become a sepulchral space until the deposition of the last burials in the Bell Beaker period. The cave would be used again as a storage space in the Late Bronze Age and, from that moment onwards, it would have been sporadically visited during the Iron age and throughout historic times (Morales *et al.* 2013). Recent excavations in the exterior have also revealed the existence of an Early Neolithic layer (Ie). This one is located at a space not covered by the cave's roof and, therefore, highly exposed to weathering, which generated lixiviated animal bones and pottery. Nevertheless, a charcoal remain from Laurus nobilis has been dated from 6150±30 (5209-5002 cal BC.)(Oms *et al.* 2016a).

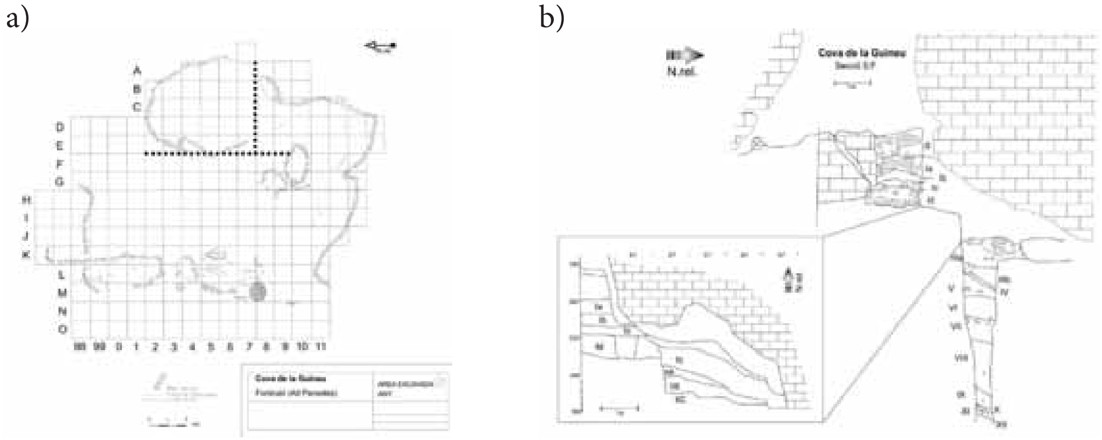


Figure 66: a) plan from the interior and exterior of the cave, b) stratigraphic profile depicting the sampled layer IIC. Reproduced from Morales *et al.* 2013.

So far, it seems the cave human occupations in the sixth millennia would have used the cavity as a place to store food and provide shelter for a mainly ovicaprine heard (Allué 2003a). The ceramic materials from the Early Neolithic have a diverse origin within the site. Some originated from the 70's excavations in sector II, where a relatively thin sedimentation ratio is assumed to be present, and others from modern day fieldwork in sector Ie. Moreover although materials from level Ie are scarce, this sector presents a specific stratigraphy which help place pottery fragments in specific time ranges (Figure 66).

5.5.3 Archaeological materials

5.5.3.1 Pottery remains

Regarding the pottery characteristics. research by Oms (2014) indicates a total amount of 43 decorated fragments belonging to at least 32 possible pots modelled in clays presenting mica, calcite and quartz. Exterior surfaces were either polished or smoothed and the vessels were fired in an atmosphere without control of the oxygen input (resulting in a mix of orange and black colours). Amongst the shapes detected in the assemblage there were eight big shperic/cilindric vases, three middle sized hemispheric and two middle sized necked vessels. The decoration practiced in these vessels included 9 simple and complex cardial motives in mainly horizontal themes (Oms *et al.* 2016a).

5.5.3.2 Lithic remains

The lithic assemblage in layer Ie contained mainly flint blade fragments and some remains of knapping activities. The range of flint pieces detected suggested the authors that lithic tools would have been used in the cave but produced elsewhere. This would not be the case of quartz pieces, which could have been locally prepared. Other tools in the site were small spoons or spatulas and one piercer made of bone and teeth (Oms *et al.* 2016a).

5.5.3.3 Animal remains

Archaeozoological analyses of animal bones recovered from layer Ie taxonomically identified 27 out of the total 110 remains. The main domestic species in the assemblages are *Ovis aries/Capra hircus* (NR=10) followed by *Bos taurus* (NR=4 MNI=2) and *Sus domesticus* (NR=2 MNI=2), which comprise 59% of the total. Detected wild species were *Oryctolagus cuniculus* (NR=8), *Cervus elaphus* (NR=2 MNI=1), *Felix silvestris* (NR=1, NMI=1) and *Equus caballus* (NR=1 MNI=1) (Oms *et al.* 2016a). These data indicate the predominance of ovicaprines in the site but also highlights the importance of wild taxa, which adds up to 41% of the total. Moreover, to facilitate comparison with organic residues, it can be stated that ruminant animals seem to be 60% of the assemblage, cecotrophs would be 29% and non-ruminants would be 11%. As reported by researchers (Oms *et al.* 2016a), 16% of the remains in the assemblage contained some degree of thermic alteration. Only one piece of seashell has been recovered in the Early Neolithic layers in the site. This low amount is coherent with the fact that Cova de la Guineu is 30km away from the sea (Oms *et al.* 2016a).

5.5.3.4 Plant remains

No published data on plant remains from the early neolithic layers in Cova de la Guineu beyond the charcoal used to obtain a ^{14}C date in layer Ie could be located in the literature.

5.5.4 Sampling and analytical techniques

Organic residue analyses from sherds found in the base of layer IIC (Morales *et al.* 2013) may provide the opportunity to gain new knowledge about the uses pottery had in Cova de la Guineu. To this end, 13 sherds were selected, including eight rims, three walls and two necks. The fragments presented orange to dark brown colourations, which are coherent with the firing techniques available in the Early Neolithic. Eight samples presented cardial impressions, seven samples had appliqués in its exterior surface and, in sample CG27, impressions were placed in the rim. Vessel 3 is of significant importance given that it preserved the shape of a jar with a globular body, a straight neck and a handle starting in the rim (Table 39).

Lab ID	Vessel	Context	Sherd id	Shape	Firing	Deco	Deco	Weight (mg)
CG1	1	E4	327	Rim	Mixed	Cardial	None	101
CG2	2	D1	599	Neck	Oxidised	Cardial	None	103
CG3	3	E3	796	Rim	Oxidised	Cardial	Handle	96
CG17	17	?	802	Wall	Oxidised	Cardial	appliquée	101
CG19	19	?	?	Wall	Mixed	Cardial	Handle	99
CG20	20	?	?	Rim	Mixed	None	appliquée	105
CG26	26	?	894	Wall	Mixed	None	appliquée	128
CG27	27	?	?	Rim	Mixed	Impressed rim	appliquée	102
CGP1	4	F7	7016	Neck	Mixed	Cardial	None	100
CGP2	19	D3	707	Rim	Mixed	None	appliquée	102
CGP3	15	?	?	Rim	Oxidised	Cardial	appliquée	101
CGP4	9	A7	72	Rim	Mixed	Cardial	appliquée	100
CGP5	31	?	820	Rim	Mixed	None	None	100

Table 41: Analysed pottery samples from Cova de la Guineu.

Around 1g of pottery was drilled and lipids were extracted by an acidified methanol extraction. Gas Chromatography was used in all samples to identify specific compounds. When retention times were not indicative enough, selected residues were analysed by Gas Chromatography and Mass Spectrometry (GC-MS). Finally samples complying with GC-C-IRMS requisites were submitted for compound specific isotopic analysis. All work was performed in the ICTA laboratories.

Lab ID	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
CG1	235.2	0.3	-28.31	-27.79	0.52	Degraded non ruminant adipose fat
CG2	11.1	1.7	-26.16	-27.4	-1.25	Ruminant adipose fat
CG3	7.5	2.6	-25.33	-26.72	-1.39	Ruminant adipose fat
CG17	10.9	2.5	-25.49	-25.96	-0.47	Ruminant adipose fat
CG19	9.0	2.2	-	-	-	Degraded archaeological fat
CG20	9.2	2.2	-	-	-	Degraded archaeological fat
CG26	13.0	1.0	-26	-27	-0.96	Ruminant adipose fat
CG27	156.3	0.4	-27.39	-28.17	-0.78	Ruminant adipose fat
CGP1	2.9	2.7	-	-	-	Non significant
CGP2	7.6	2.4	-	-	-	Degraded archaeological fat
CGP3	74.0	0.8	-27.24	-26.65	0.59	Degraded non ruminant adipose fat
CGP4	14.7	4.1	-	-	-	Degraded archaeological fat
CGP5	13.4	4.1	-	-	-	Degraded archaeological fat

Table 42: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

5.5.5 Results and discussion

After a broad quantification using the previously mentioned standards, significant amounts of lipids were detected in 12 out of the 13 analysed samples (92%) (Table 40). Except from two outliers, the amounts of recovered fats averaged around 15µg·g^{-1}. In the case of samples CG27 and CG1, these pots contained more than ten times the mean amount of residues for the rest of the assemblage (see table 40). If it is assumed that postdepositional processes had roughly the same impact into lipid preservation across layer IIC, vessels 1 and 17 could have been used in more activities involving high quantities of fats. Nevertheless, although the pieces with the highest lipid yields were rims, it needs to be taken into account that walls and necks did also present significant amounts of fat. Furthermore, the presence of cardial impressions did not seem to be associated with either higher or lower amounts of residues (Mann Withney U = 15 n=13 p = 0.69).

The recovered residues were mainly composed of free saturated fatty acids ranging from 9 to 30 carbon atoms in its chain. The amount of cases (CG1, CG3, CG26, CG27 and CGP5) where nonanoic acid (C9:0) was detected (41%) was significantly higher when compared with residues in other archaeological sites (ex: Guixeres de Vilobí, La Serreta), where the shortest fatty acids usually incorporate at least two more carbon atoms. Nevertheless, these are only minor constituents of the overall residue and could be easily explained by the presence of slightly different postdepositional effects. As it will be attested by isotopic values, short chained fatty acids in these samples did not seem to originate from dairy triacylglyerols. Oxidation products in the shape of keto acids and dicarboxilic acids such as azelaic acid (CG1, CG2, and CG27) suggest unsaturated molecules were also affected by postdepositional effects. The presence of alkanes and phthalate plasticisers might be explained by the use of plastic bags to store the sherds after excavation. The absence of specific biomarkers for plant residues such as odd over even quantities of alkanes and specific sterols and the fact that (in samples CG1, CG2, CG3, CG17, CG19, CG20, CG26, CG27, CGP2 and CGP3) palmitic and stearic acids seem to present roughly the same abundance suggests any major inputs of plant fats should be rejected. Nevertheless, this fatty acid profile is not incompatible with a residue probably originated in the hydrolysis of animal triacylglycerols. This is less clear for samples CGP4 and CGP5, with a P/S value around 4. These high values have been previously reported as associated with residues from a plant origin (Copley *et al.* 2005e, Dunne *et al.* 2016), nevertheless no further data supporting this evidence prevent a clear identification of plant residues in Cova de la Guineu. To further characterise vessels possibly containing significant amounts of animal fats, seven samples were submitted to compound-specific isotopic analyses (Figure 67). Measurements from palmitic and stearic acids were plotted and compared with modern authentic reference materials.

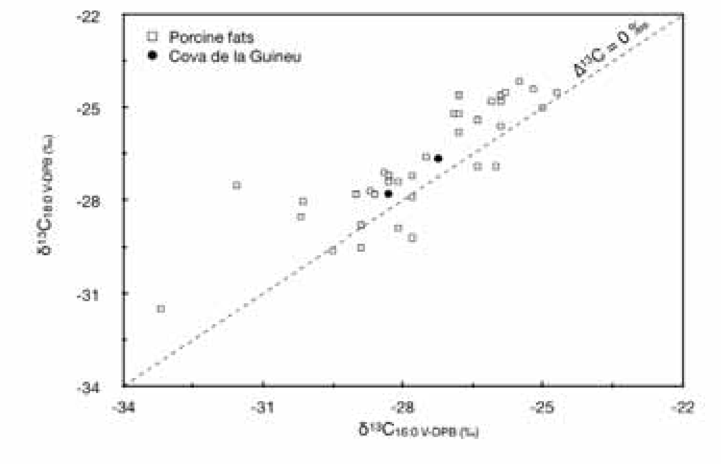


Figure 67: Comparison of δ^{13}C16:0‰ and δ^{13}C18:0‰ non-ruminant modern reference values with selected samples CG1 CGP3 from Cova de la Guineu. See figure 56 for reference data.

Samples CG1, CG2, CG3, CG17, CG26, CG27 and CGP3 presented isotopic values coherent with the results obtained from other Early Neolithic sites in this study. In two cases (CG1 and CG3), given that Palmitic acid was more enriched in δ^{13}C than stearic acid, values could be associated with non-ruminant fats. Non-ruminant species detected in the site were Felis silvestris and Sus domesticus. In the case of the wild cat, the fact that the only remain was a perforated tooth used in a necklace suggests the most probable origin for these non-ruminant residues

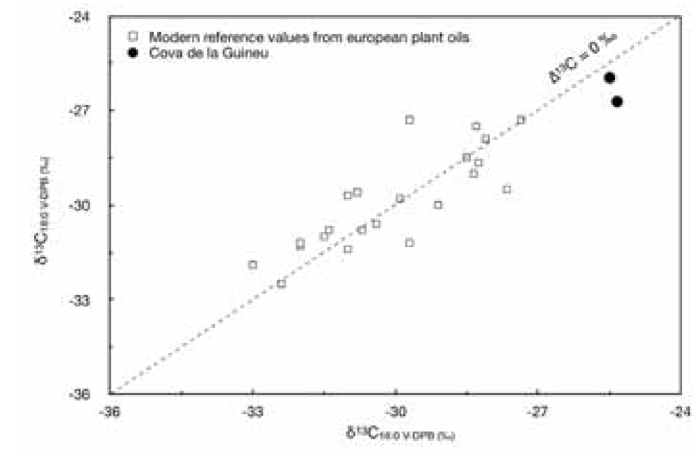


Figure 68: Comparison of δ^{13}C16:0‰ and δ^{13}C18:0‰ plant oil modern reference values with selected samples CGP4 and CGP5 from Cova de la Guineu. Reference data: Dudd 1999, Copley *et al.* 2001, Spangenberg and Ogrinc 2001, Steele *et al.* 2010, Horiuchi *et al.* 2015, Lucquin *et al.* 2016b.

would be pig adipose fats. A comparison with published modern authentic pig fats (Evershed *et al.* 1997a, Copley *et al.* 2003, Spangenberg *et al.* 2006, Gregg *et al.* 2009, Horiuchi *et al.* 2015, Lucquin *et al.* 2016b, Colonese *et al.* 2017b) shows that the samples fall within the expected area for this species (Figure 68).

Samples CG17 and CG3 presented P/S ratios higher than 2. In order to completely rule out possible plant inputs and given that no specific plant biomarkers were detected in these residues, it seemed unlikely that palmitic and stearic δ^{13}C isotopic values would be similar to modern reference plant oils. Figure 69 shows that, in this case, the values from these two vessels are separated from the expected area where plant oils plot (Dudd 1999, Copley *et al.* 2001, Spangenberg and Ogrinc 2001, Steele *et al.* 2010, Horiuchi *et al.* 2015, Lucquin *et al.* 2016b) and, therefore, the residues most probably originated from ruminant adipose fats.

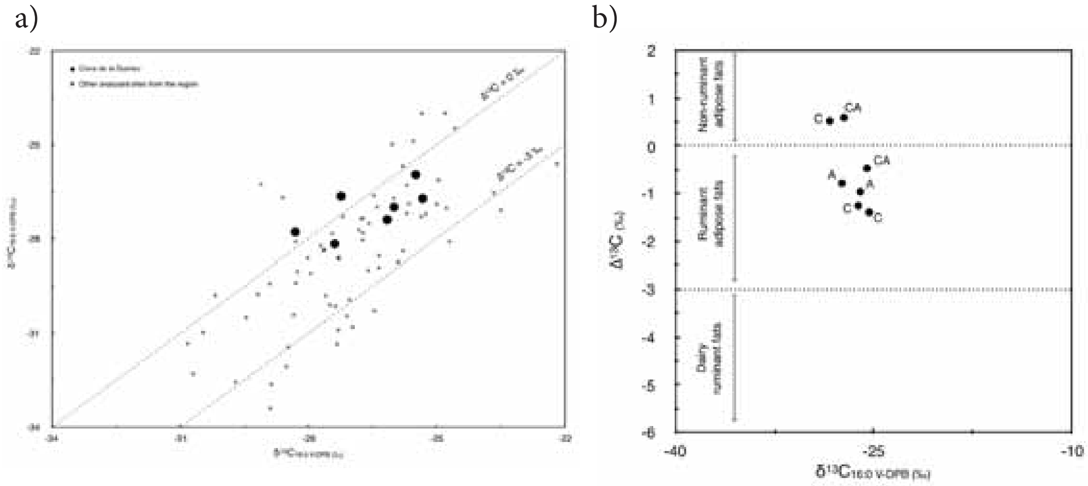


Figure 69: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

Therefore, it seems that 71% of the animal fats analysed in the assemblage would have belonged to ruminant animals while 29% could have mainly been associated with non-ruminant adipose fats, most probably Sus domesticus. These data seem to be approximately similar to the percentage of ruminant (60%) and non-ruminant (11%) bones in layer Ie in Cova de la Guineu.

Nevertheless, given that residues in vessels would result in an average from multiple uses, it is safe to assume that at least some level of mixing could have occurred. Working under this assumption, a bayesian mixing model with dairy, ruminant and non-ruminant end-members has been applied to Cova de la Guineu's results to evaluate the overall importance of each possible product.

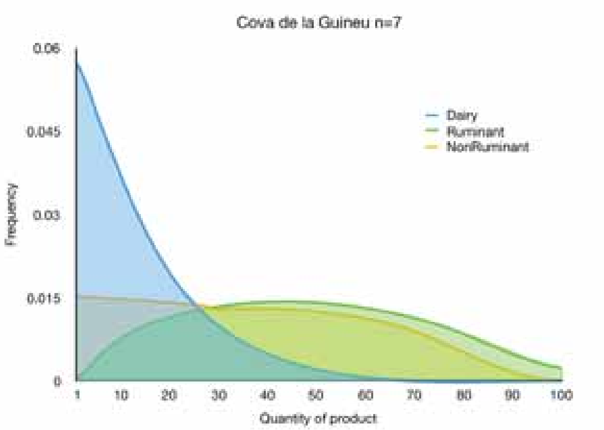


Figure 70: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all samples from Cova de la Guineu.

Results from the model (Figure 70) assessing all pottery suggest had there been any dairy products in the vessels, it is highly unlikely the amounts were significant (median=11%). Alternatively, similar quantities of ruminant and non-ruminant adipose fats might have been the two main possible sources of lipids (non-ruminant median=37%, ruminant median=46%). This effect might be explained by the absence of $\Delta^{13}C$ values lower than -1.5 and the presence of two samples above 0, which the model incorporates as possible mixtures of ruminant and non-ruminant adipose fats (all cases), possible non-ruminant exclusive (CG1, CGP3) and possible ruminant exclusive (GC2, CG3, CG17, CG26 and CG27).

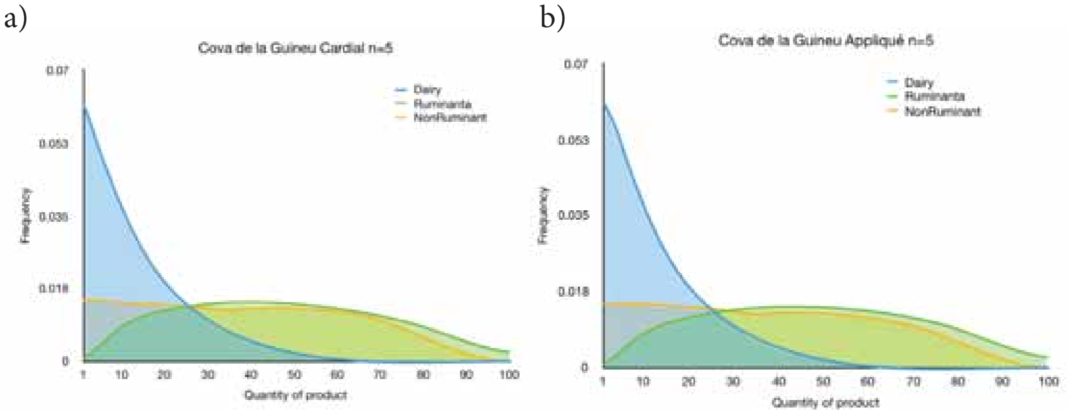


Figure 71: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in samples presenting a) cardial impressions and b) appliqués.

In more detail, most of the analysed sherds from Cova de la Guineu were decorated with cardial motives and/or presented an appliquée. To assess wether the presence of these features could be linked with the contained products, the results of the bayesian mixing model were split between these two categories.

It does not seem that the model (Figure 71) would indicate any major differences in content between these two types of decorative techniques. Both follow the general trend in the site of low to negligible dairy and significant ruminant and non-ruminant. The subtraction of each percentile probability (Figure 72) did not seem to suggest the existence of any minor differences. The high instability of the probability curves indicates any possible differences are close to the accuracy of the model itself and, therefore, do not correspond to significant changes in vessel use. In conclusion, the applied bayesian mixing model does not support a differential vessel use between cardial and appliquée decorations in Cova de la Guineu. This phenomenon might be explained by the fact that both cardial decorations and appliqués are present in three of the compared vessels, thus biasing making any statistical measure of the differences. In any case, it shows a different trend from what was observed in La Serreta.

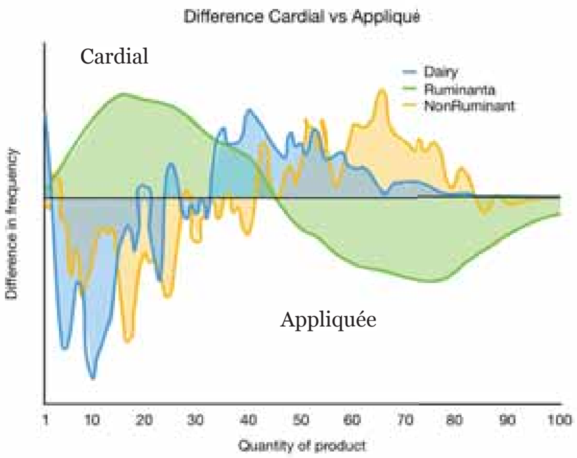


Figure 72: Comparison of the modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in sample containing cardial decorations and appliqués.

5.5.6 Conclusion

Therefore, it could be interpreted that, for this site, both types of animals could have been processed using culinary techniques involving pottery. Nevertheless, it should be stated that other possible residue types such as dairy products and marine oils could not be detected in the analysed vessels. Although the distance from the sea would suggest marine products would have been difficult to access, one piece of seashell found amongst the Early Neolithic layers in the cave indicated that a minor arrival of marine products could still be possible. If that was the case, organic residue analyses would indicate that the processing marine products would have not been associated with pottery. Although molecules associated with thermal alterations such as long chain ketones (Raven *et al.* 1997) or cyclic fatty acids (Cramp and Evershed 2014) were not detected, sample CGP5 presented trace amounts of anthracenes and napthalenes. These molecules could originate from wood combustion and they could have been incorporated after multiple periods of exposure to open fires (Papakosta *et al.* 2019, Oudemans and Boon, 1991 and Skibo 2013). Nevertheless, it is not clear when these combustion residues could have infiltrated the sherd. The presence of foumiere-type features in layer II (Bergadà *et al.* 2003) suggests post-depositional contamination is a factor that can not be completely ruled out. In any case, animal fats would have been easily incorporated into the clay matrix in liquid form.

Consequently, it is to be expected that those vessels containing significant amounts of animal fats could have been subjected to at least minor heat while containing some type of animal product. Finally, one of the most significant pieces in the Cova de la Guineu is vessel 3. This jug contained, according to residue analyses, degraded ruminant animal fats. Explaining the presence of this product in a shape usually associated with serving liquids might prove challenging. One of the possibilities might be it used to contain some type of broth or liquid substance resulting from the processing of said ruminant adipose fat (a sauce?). Sadly, there is a severe scarcity of evidence to support choosing one interpretation over the other.

6 South and Ebro valley

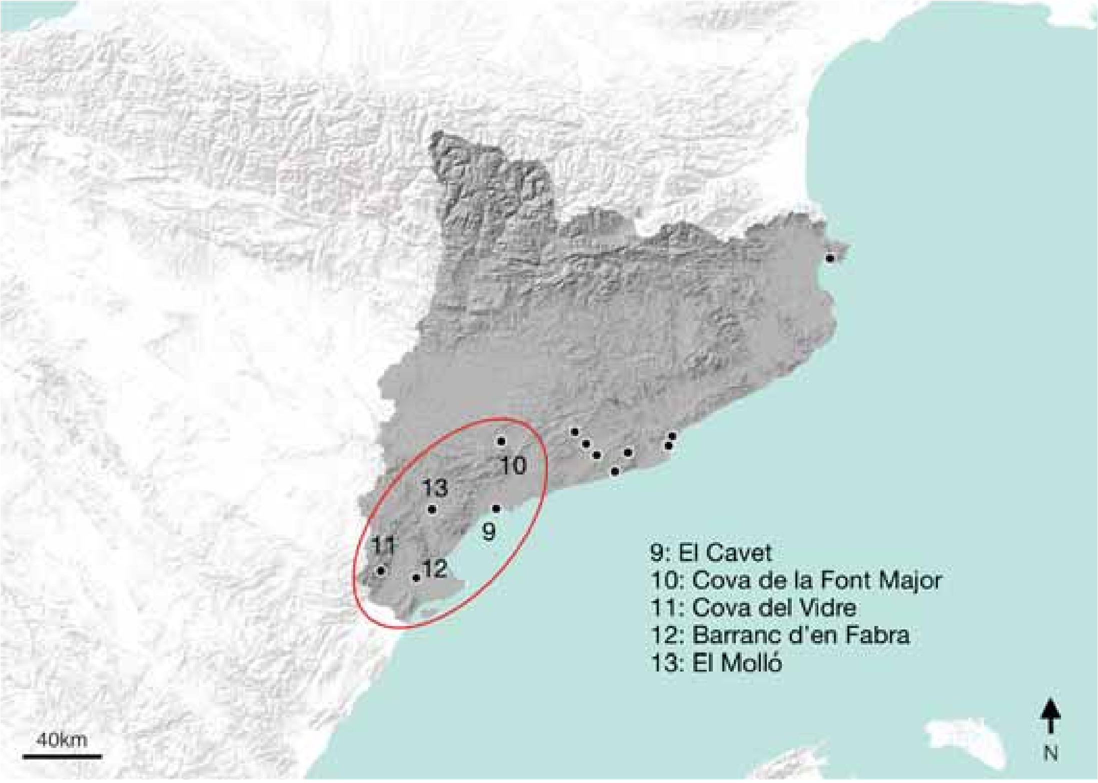


Figure 73: Map of the studied site in the South and the Ebro valley.

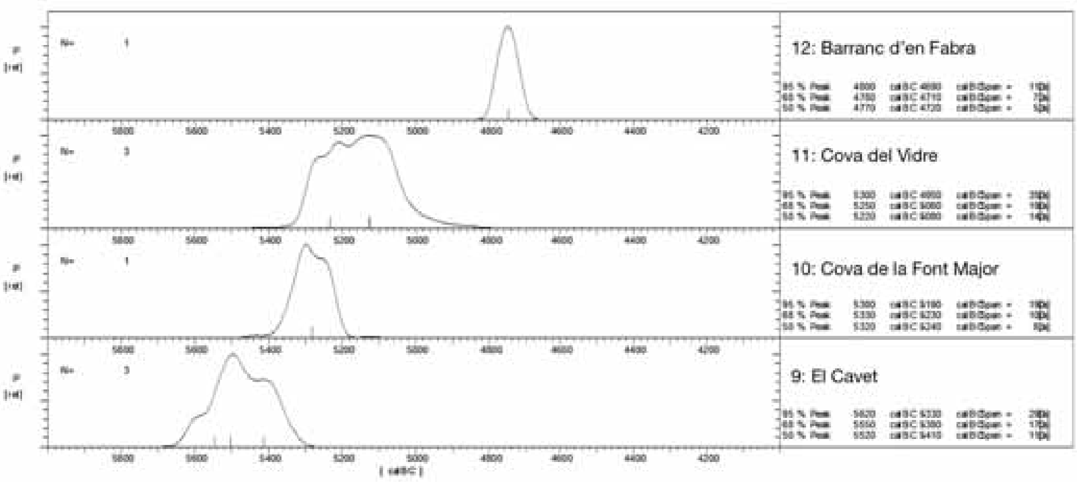


Figure 74: Calibrated ^{14}C dates from Early Neolithic contexts in the South and the Ebro valley. Software: Cal Pal v1.5

6.1 El Cavet

6.1.1 Introduction

El Cavet is an open air archaeological site in the village of Cambrils, around 20 meters above the sea level and 1.2km from the modern seashore. Since the 1980's, development work related with the urbanisation of the area and contract archaeological interventions revealed the presence of a significant archaeological site around 5700 m^2 wide (Martins *et al.* 2015). In 2005, the Institute of Paleoecology and Social Evolution (IPHES) programmed a regular annual excavation to study it (Fontanals 2005, Fontanals *et al.* 2008a, 2008b).

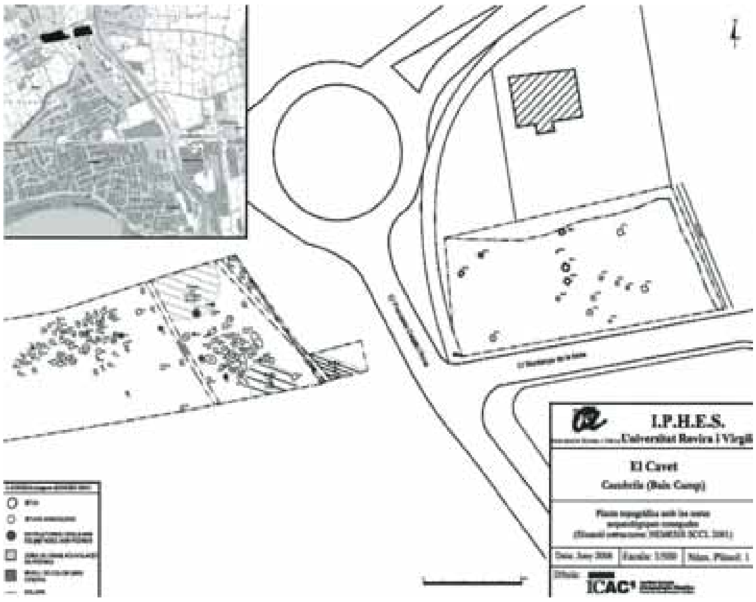


Figure 75: General plan of the structures detected in El Cavet, reproduced from Fontanals *et al.* 2008a

6.1.2 Archaeological features relevant in this study

The site is divided in three main areas (I, II and III) (Figure 75). Although area I presented no remains dating from the Early Neolithic, shallow cuts in sector II and specific strata from sector III contained artefacts and structures of significance.

A total of 15 of features were excavated in sector II. These included five which were associated with Early Neolithic occupations: UE 2012, UE 2014, UE 2016, UE 2007 and UE 2009. They were circular pits with concave walls, flat bases and globular in section. Rims oscillated between 100 and 150cm of diameter and no more than 65cm of depth (Fontanals 2005, Fontanals *et al.* 2008b). Given the pit's morphology, the researchers interpret these features could have been used as silos, which were later subject to the dump of domestic debris. The excavation detected that the feature's interiors had been refilled by a sandy and very compact sediment with limestone pebbles, sandstone, quartz and granite from a nearby water stream (Riera de Maspujols) (Fontanals *et al.* 2008b). The recovered materials were mainly pottery, lithic tools, marine and terrestrial seashells, charred seeds and charcoal.

Furthermore, in sector III, an area with two superimposed archaeological layers has been shown to contain Early Neolithic occupations. Lithic industry, thermally altered blocks and pottery with cardial decorations were found in UE300 and the negative structure named UE117.

Feature 2012 presented at least two differentiated refilling layers (2012.1 and 2012.2) containing lithic tools, pottery, charcoal and charred seeds. A sample of *Triticum aestivum/durum* was dated from 6536±36 BP (Oms 2014).

Feature 2014 underwent a different postdepositional process. A slow refilling time was inferred by the researchers due to the presence of sediment originating in a partial pit collapse between two different refilling layers. Charcoal from Quercus sp. was dated from 6590±60 BP (Fontanals *et al.* 2008a) and *Triticum aestivum/durum* seeds were dated from 6440±40 BP (Oms 2014).

Silo 2016 presented only one refilling layer containing 12 pottery sherds (14% of the period's total assemblage) fired under mixed atmospheres. No ^{14}C dates have been acquired from this silo to date (Fontanals *et al.* 2005).

Finally, features 2007 and 2009, although ascribed to the same period, contained highly weathered materials preventing an accurate chronological attribution (Oms 2014).

6.1.3 Archaeological materials

6.1.3.1 Pottery remains

Silo 2012 contained significant ceramic material although it was only found after the first 15cm of depth (Fontanals *et al.* 2008a). At least 61 sherds (70% of the period's total assemblage) belonging to 18 different vases were recovered in the 2012.2 layer. Some of these were straight walled storage containers, other were middle sized pots with ovoid shapes and a neck and subespheric pots and, in minor quantities, small vessels with open rims. All pottery had been fired under highly variable atmospheric conditions as recorded by the mix of oxidising and reducing chemical reactions across the vessels. A percentage of the recovered pottery was decorated per the stylistic conventions for the cardial Early Neolithic, this being the use of plain appliqués and seashell "baroque" impressions (Oms 2017a).

Silo 2014 contained 11 sherds (13% of the period's total assemblage) which belonged to at least 5 different vessels. One of them featured some cardial decorations and another had straight walls and large dimensions. These two originated from context 2014.2. The first deposit (2014.1) contained the remaining 3 vessels which were characterised by straight walls and rims, flat or rounded lips and middle sized vases with ovoid or subesferic shapes.

In silo 2016, several decorated fragments including one presenting a horizontal motive crossed by a vertical cone was used by researchers to attribute the assemblage to an undetermined more recent phase in the Early Neolithic (Fontanals *et al.* 2008a, 2008b).

6.1.3.2 Lithic tools

Around 150 lithic remains and 14 finished artefacts evidenced the production of lithic tools in the site by using mainly flint but also other materials such as quartz, jasper, agate and caliche. A red to garnet residue found in many lithic pieces is also present as a thick impregnation in the interior wall of a potsherd. According to Fontanals *et al.* (2008a) analysis of said residue is currently underway.

6.1.3.3 Animal remains

Analysis of preserved organic remains shed light into the socioeconomic activities performed by the human groups present at El Cavet. Marine seashells in silos from Sector II were mainly gastropods (77%) and bivalves (23%). The bulk of the remains were found in silo 2012, mainly in the 2012.2 stratigraphic units. Other silos contained a diversified range of species (2014.2 and 2016.2). Out of a wide range of 9 gastropod species, one *Nassarius mutabilis* had been artificially perforated and one *Charmonioa lampas* presented polishing from anthropic origin in the symphonial face. Bivalves were mainly *Acanthocardia tuberculata* (19 MNI), *Arca noae* (3 remains) and, a fragmented *Glycimeris* sp. (Fontanals *et al.* 2008a).

6.1.3.4 Plant remains

Charcoal from the 2004 to the 2006 campaigns (Allué 2003b) belonged to *Pinus halepensis*, *Juniperus* sp, *Olea europaea*, *Quercus ilex/coccifera*, *Quercus* sp. and *Erica arborea* amongst others. The presence of these species brought the researchers to interpret that the site's vegetal environment could have been composed by Mediterranean garrigue vegetation. Regrettably, no animal bones have been found in the studied contexts so far, probably due to the site aggressive sedimentary conditions.

Given the difficulty to recover interpretable faunal remains due to high soil acidity, the selection of vessels from all the structures and contexts associated to date with the Early Neolithic might be able to bring some information on the possible animal products consumed in the site. Moreover, a site so close to the sea could provide an excellent context to evaluate the extent of the transformation of marine oily products in pottery.

6.1.4 Sampling and analytical techniques

A range of 22 pottery vessels were selected for organic residue analysis (Table 40). In sector II, the majority (n=13) were recovered from feature 2012 and from layer 2, one sample was taken from feature 2014 layer 1 and another one from feature 2016 layer 1. In sector III, 4 samples were selected from stratigraphic unit 300 and two from the second layer in the feature, 117. Finally one sample from the REM context was also studied. This assemblage was mainly composed of rims (n= 15) but also included walls (n=4), a neck (CV25), and a fragment close to the base (CVP6). Sherds 496 (CVP5) and 521 (CVP6) belonged to the same vessel and offered the opportunity to compare the composition of residues found in different parts of the vessel profile. Regarding the coloration, which could be related to the firing techniques, 14 samples presented a gradient from brown to orange, one was consistently dark and six were mainly a combination of yellow to orange colours. Around 54% of the assemblage (n=12) were plain sherds, but two fragments were decorated with impressions, one was incised and 6 fragments presented plastic applications in the shape of appliqués, handles and one short strip. The selection of samples included a range of possible vessel functionalities depending on the presence of gripping implements and sizes.

Lab ID	Arch context	Date	Vessel Numb	Sherd id	Vessel part	Colour	Decoration	Depth	Sampled Weight (mg)
CV114	2012.2	2006	-	114	Rim	Mixed	None	89	83
CV164	2012.2	2006	-	164	Rim	Mixed	None	94	102
CVREM	REM	?	V33	2414	Rim	Reduced	Incised	?	104
CV25	G73	2011	V40	25	Neck	Mixed	Cardial	1084	98
CV280	2012.2	2006	-	280	Rim	Oxidised	None	104	92
CV3003	300/F70	2008	-	3	Rim	Mixed	None	1091	103
CV36	300/G70	2008	-	36	Rim	Oxidised	None	1103	100
CV37	300/D7	2008	V37	10	Rim	Mixed	Cardial	1094	92
CV438	2012.2	2006	-	438	Rim	Mixed	None	121	100
CV52	2012.2	2006	V52	172	Rim	Mixed	Appliquée	93	105
CV522	2012.2	2006	-	522	Wall	Mixed	None	120	111
CV53	2012.2	2006	V53	433	Rim	Oxidised	Appliquée	125	101
CV56	2012.2	2006	V56	462	Wall	Oxidised	Appliquée	110	103
CV60	117.2	2009	-	60	Rim	Mixed	None	1144	73
CV66	2012.2	2006	-	66	Rim	Mixed	Strip	?	111
CVN56	117.2	2009	-	56	Wall	Oxidised	Handle	1143	100
CVP1	2012.2	2006	-	228	Wall	Oxidised	Handle	98	103
CVP2	2016.1	2005	-	68	Rim	Mixed	None	70	104
CVP3	2012.2	2006	-	440	Rim	Mixed	Appliquée	122	99
CVP4	2014.1	2005	-	90	Rim	Mixed	None	101	100
CVP5	2012.2	2006	-	496	Rim	Mixed	None	119	101
CVP6	2012.2	2006	-	521	Base	Oxidised	None	122	103

Table 43: Analysed samples from El Cavet.

All samples were extracted using the acidified methanol technique and submitted to GC analysis. When retention times were not sufficient to identify significant peaks, selected samples were analysed by GC-MS and the acquired mass spectra was compared to the NIST2.0 library. Samples yielding a TLE higher than $5\mu g \cdot g^{-1}$, presenting a P/S ratio coherent with animal fats and absence of UCM or problematical background noise were analysed via GC-C-IRMS. All work was performed in the ICTA laboratories.

6.1.5 Results and discussion

Alternatively to other archaeological sites included in the study, a significantly higher percentage (37%, n=8) of the vessels sampled for organic residue analysis in El Cavet yielded non-significant amounts of lipids. Although roughly 60% of the sherds originated from feature 2012, around 75% of the non-significant residues (n=6) were located there (Table 41). The two remaining negative results were vessels from stratigraphic unit 300. Therefore, although eight sherds contained no significant data, sufficient quantities of lipids could be extracted in vessels from all the studied features. Overall, lipid quantities were also lower than other studied sites. The highest yield for El Cavet was only 66.6µg·g^{-1} and around 60% of the analysed vessels contained lipid quantities lower than 10µg·g-1. Despite more severe than average postdepositional processes could have challenged the preservation of lipids, it should also be taken into account the possibility that some of the studied sherds were never exposed to significant quantities of archaeological fats in the first place.

LAB id	TLE	P/S	δ1³C16:0‰	δ¹³C18:0‰	Δ¹³C‰	Interpretation
CV114	5.0	2.4	-	-	-	Non Significant Residue
CV164	5.1	1.9	-	-	-	Degraded animal fat
CVREM	30.9	1.1	-30.46	-30.97	-0.51	Degraded ruminant adipose fat
CV25	6.1	1.9	-	-	-	Degraded animal fat
CV280	4.4	1.8	-	-	-	Non Significant Residue
CV300/3	10.9	5.2	-28.91	-29.42	-0.51	Possible ruminant adipose
CV36	1.5	1.9	-	-	-	Non Significant Residue
CV37	5.0	3.0	-	-	-	Non Significant Residue
CV438	4.2	1.3	-	-	-	Non Significant Residue
CV52	nd	nd	-	-	-	Non significant Residue
CV522	6.8	1.3	-	-	-	Degraded animal fat
CV53	6.3	1.6	-28.26	-29.04	-0.78	Degraded ruminant adipose fat
CV56	5.7	1.3	-	-	-	Degraded animal fat
CV60	14.1	0.9	-	-	-	Degraded animal fat
CV66	2.3	2.3	-	-	-	Non Significant Residue
CVN56	11.1	3.6	-29.19	-29.76	-0.57	Possible ruminant adipose
CVP1	1.0	3.2	-	-	-	Non Significant Residue
CVP2	19.9	2.0	-26.77	-27.38	-0.61	Degraded ruminant adipose fat
CVP3	42.5	1.0	-28.02	-28.62	-0.6	Degraded ruminant adipose fat
CVP4	6.8	2.4	-	-	-	Degraded animal fat
CVP5	66.6	0.7	-28.52	-32.06	-3.55	Degraded ruminant dairy fat
CVP6	13.3	2.2	-	-	-	Degraded animal fat

Table 44: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in µg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio.

Roughly all samples with significant amounts of lipids presented a series of long chained alkanes and phthalate plasticisers resulting from possible contamination by plastic bags. Free fatty acids were mainly saturated and ranged from 9 to 28 carbons in the acyl moiety. The most abundant were consistently Palmitic (C16:0) and Stearic acids (C18:0), although C12:0, C14:0 and C20:0 were also usually present across the assemblage in minor quantities. Unsaturated fatty acids were mainly oleic (C18:1) and palmitoleic (C16:1) although in some cases (ex: samples CVP5, CVP6) azelaic acid and a series of -oxo octadecanoic acids indicated the survival of the oxidation products of previous unsaturated compounds. The relevance of the nonanedioic acid peak suggests that double bonds were mainly in Δ^9 positions, which would be expected if oleic acid had been the main source of oxidation products. Although oleic acid is significantly abundant in plant oils, it is also a main compound of animal triacylglycerides and, therefore, the presence of said oxidation products should not be used as a marker for a possible plant input.

Additionally, sample CVP2 presented two diterpenoids associated with pine resins. These, dehydroabietic acid and 7-oxo dehydroabietic acid, could be coherent with minimal inputs of degraded resins. Such minor amounts of diterpenoids make it difficult to assess with certainty wether they had an archaeological origin. Nevertheless, had that been the case, rather than waterproofing the vessel inner surface with pure resin, the origin of these compounds could be associated with the transformation of plant products containing minor amounts of resins. Given the difficulty to provide a solid interpretation from these molecules, their presence in El Cavet should not be assumed as evidence for the manipulation of resin during the Early Neolithic, but as a possibility that might or might not be confirmed provided that further evidence supports or rejects this hypothesis in the future.

Apart from CVP2, no plant specific biomolecules such as odd chained alkanes, sitosterol, wax esters or resin triterpenoids were detected. This would imply that the possibility of a major contribution of plant material to the residues should be rejected. In some positive samples (CV300/3 and CVN56), the presence of up to 5 times more palmitic than stearic acid would not be incoherent with the degradation of plant triacylglicerols (Copley *et al.* 2005e and Dunne *et al.* 2016), but the aforementioned absence of significant plant biomarkers prevents its identification as this type of material. Alternatively, marine products have been shown to contain high relative abundances of palmitic to stearic acids. Nevertheless, no specific biomarkers for aquatic resources (isoprenoic fatty acids, ω-(o-alkyphenyl)alkanoic acids and very long chain unsaturated fatty acids amongst others) were detected either. In consequence, the biomolecular study of samples CV300/3 and CVN56 indicates the presence of significant amounts of degraded unspecified fats possibly originating from pottery use. Alternatively, in roughly 85% of the positive samples the ratio of palmitic to stearic acids was between 1 and 2.5. Given such free fatty acid profiles the possibility that these residues originated from animal triacylglycerides can not be refused.

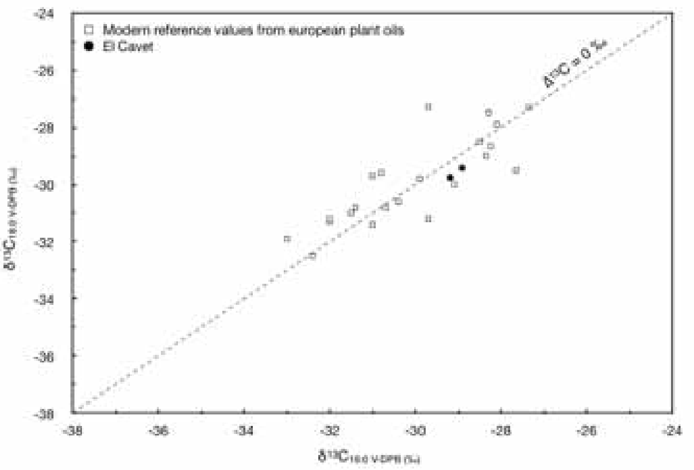


Figure 76: Comparison of δ13C16:0‰ and δ13C18:0‰ values from modern reference plant oils with samples CV300/3 and CVN56 from El Cavet. See figure 68 for reference values.

A range of isotopic and biomolecular markers were studied to further evaluate the presence and type of possible degraded animal fats present in pottery from El Cavet. Phytanic acid is the result of the bacterial oxidation and hydrogenation of phytol, the alcohol moiety in chlorophyll. Given that it can be transmitted through terrestrial food chains (Lucquin *et al.* 2016a), it has been observed in the triglycerides of ruminant adipose tissue (Lough 1977), animal bone (Colonese *et al.* 2015), butterfat (Sonneveld *et al.* 1962) and marine fats and oils (Ackman and Hooper 1968). Furthermore, their presence in pottery has been used to help the interpretation of marine and/or ruminant organic residues (Lucquin *et al.* 2018). Nevertheless, although experimental research suggests incorporation of fatty acid contamination from bacteria or the migration of soil lipids during decay in the burial environment is considered to be minor (Dudd 1999, Heron *et al.* 1991) post-depositional bacterial degradation of chlorophyll as a result of the introduction of plant material in the vessel should not be rejected. Furthermore, omnivore non-ruminant animals feeding on matter containing phytanic acid such as waste meat or whey could also incorporate into depot fats (Dudd 1999). In consequence, when no further study of its diastereoisomers is available (Lucquin *et al.* 2016a), the interpretation of this molecule in organic residues must always be treated with caution.

Branched chain fatty acids are major constituents of microbial membranes and they are also fermentation end products used as metabolic substrates in the rumen (Dudd 1999). Assuming the incorporation of bacterial constituents from soil to be a minor factor explaining well protected lipidic residues in pottery (Dudd 1999, Heron *et al.* 1991), the relative amount of branched-chain fatty acids has been used as a supporting criteria (Regert 2011) for the presence of ruminant related products in pottery organic residues. The ratio of the iso and anteiso branched heptadecanoic acids (iC17:0br + aC17:0br) in relation to stearic acid (C18:0) has been proposed as a proxy to evaluate said intensity. It is assumed that the similar molecular weights and structures these three compounds share, would make diagenetic influences minimal (Dudd *et al.* 1999). The validation of this marker has so far been performed in archaeological pottery where both biomolecular and isotopic data are available. Both Dudd *et al.* (1999) and Hjulström *et al.* (2008) report the absence of non-ruminant $\Delta^{13}C$ values in samples with C17:0br/C18:0 ratios higher than 0.02. In absence of further studies on modern authentic reference fats, this ratio has been considered only a complementary indicator for the possible presence of ruminant-related products. In any case, further supporting evidence from isotopic analyses is always needed.

In El Cavet, samples CVP5, CVP2 and CVP3 present C17:0br/C18:0 ratios all slightly above 0.02 (see Table 42). In the case of samples CVP5 and CVP3, although isotopic data is needed for confirmation, these values could still not be incompatible with the presence of non-ruminant fats. Nevertheless a 0.08 value in CVP2 strongly suggests the presence of a significant amount of ruminant fats in this vessel.

Sample	**C17:0br/C18:0**
CVP5	0.025
CVP2	0.080
CVP3	0.029

Table 45: C17:0br/C18:0 values from samples in El Cavet.

To obtain further data on the origin of the palmitic and stearic acids and after taking into account the low preservation rate and the need to remove samples with significant background noise and unresolved complex mixtures from the study, seven samples were submitted for compound-specific isotopic analysis. In this case, two samples with Palmitic/Stearic ratios above 3 were also included with the aim to obtain the fullest isotopic picture possible.

Isotopic values for samples with similar amounts of palmitic and stearic acids do not plot near non-ruminant, plant or marine reference values, thus suggesting

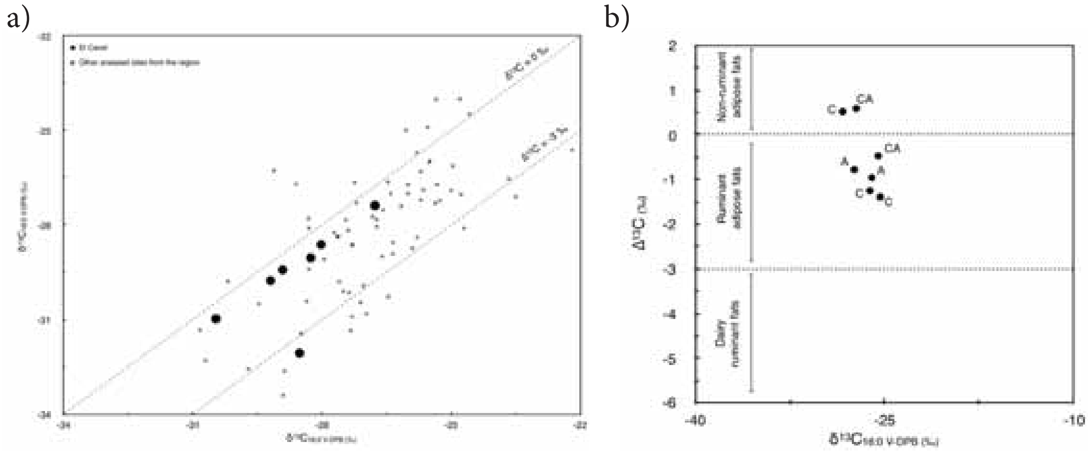


Figure 77: Results from the compound specific isotopic studies. a) plot presenting the δ13C:18:0 and δ13C:16:0 values b) plot presenting the Δ13C and δ13C:16:0 values.

a significant presence of said products in the residue can be rejected. Moreover, isotopic values for samples CVP5, CVP3 and CVP2 are coherent with C17:0br/C18:0 values in pointing at possible significant quantities of ruminant fats. Alternatively, these samples all plot near modern reference ruminant adipose fats and, in one case, ruminant dairy fats (Figure 77).

In the case of samples CV300/3 and CVN56, where palmitic acid was significantly more abundant than stearic acid, values are similar to references from modern ruminant adipose fats. Nevertheless, a comparison with published plant modern references shows that the two archaeological samples also plot close to the edge of the distribution (Figure 76). Given these values, the option that minor amounts of plant residues were present in the two analysed vessels should not be fully rejected. The absence of specific plant biomarkers could be explained by the extensive degradation process affecting the preservation of organic matter in El Cavet as a mixture of plant and animal degraded triglycerids is not inchoerent with the fatty acid profile recovered in these two samples. In any case, the possible presence of plant residues should not prevent the evaluation of the possible animal fats which could have generated the observed isotopic values.

Following this line of thought, the presence of possible mixtures has to be assessed in all cases. To this end, a bayesian mixing model including dairy, ruminant and non-ruminant end members was applied to the compound specific isotopic values from El Cavet. Given the absence of specific plant and marine biomarkers in the residues, these two products were not taken into account.

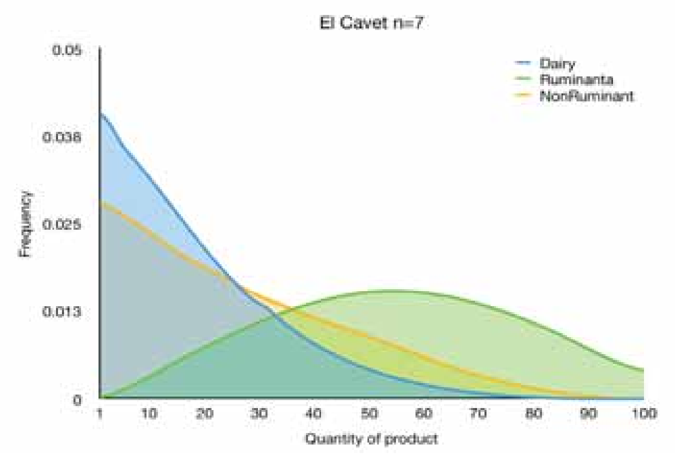


Figure 78: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from El Cavet.

Overall, model results (Figure 78) suggests the possible presence of several types of mixtures with the absence of significantly high quantities of dairy products (median = 15%) and possibly slightly higher quantities of non-ruminant fats (median= 22%). Ruminants could have most probably been the main component in the mixtures with a median around 54%. This result seems to be caused by the spread of values presenting almost equal $\Delta^{13}C$ values but with a variation of around 3‰ in the $\delta^{13}C16:0$ and $\delta^{13}C18:0$ isotopic ratios. Moreover, bearing in mind the small sample size neither vessel decoration nor sampling place within the profile (rim, wall, neck) nor the cooking atmosphere seemed to present any significant differences in use. In consequence, the model is not incompatible with the minor presence of non-ruminant adipose fats although ruminant fats might have been more frequently used. Relevant differences seem to appear when samples from sector II and sector III are compared (Figure 79).

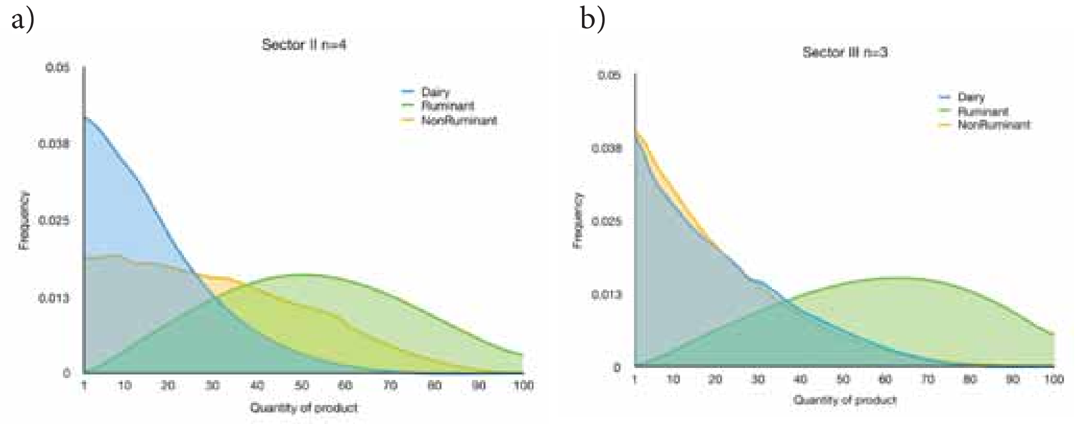


Figure 79: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in samples from a) sector II and b) sector III.

Although in both cases ruminant adipose fats could have been amongst the most probable residues present in the vessels, carcass fats from sector II presented consistently more enriched $\delta^{13}C$ values, which could also be coherent with a relative increase in the quantities of non-ruminant adipose fats (Figure 80). Nevertheless, this interpretation is conditional to the absence of isotopically enriched ruminant adipose values, a fact that can not be rejected. In consequence, although clear differences in the $\delta^{13}C16:0$ between sector II and sector III seem to suggest that vessels in these areas could have contained slightly different types of fats, the acquisition of further data that confirms the trends detected in this small sample set will be necessary.

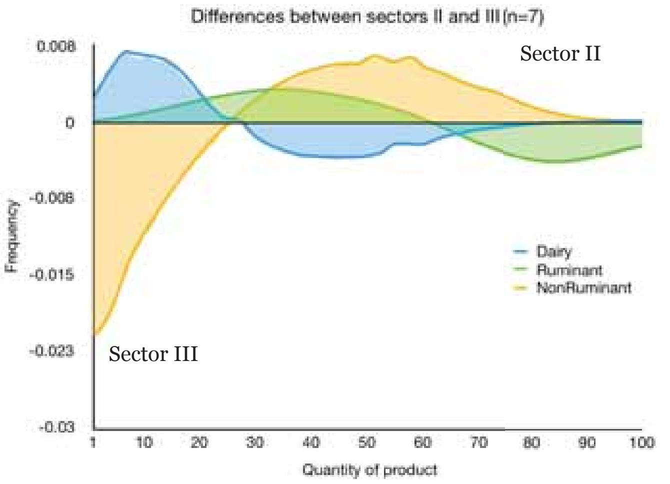


Figure 80: Comparison of modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from sectors I and II.

6.1.6 Conclusion

Overall, organic lipid residues in El Cavet could have originated from animal ruminant fats although the possibility that minimal plant inputs were also present should not be completely ruled out. The absence of biomarkers for specific marine resources in a site 1.2km away from the modern seashore where remains of several seashell species have been recovered suggests either the consumption of marine products was not significant enough, or they were cooked and consumed without the involvement of pottery. Moreover, the study also revealed the absence of isotopic values coherent with significant amounts of non-ruminant adipose fats such as pig. This phenomenon could be explained by the small amount of samples analysed. Similar to marine products, had pigs been present in the site, it is possible that they were specifically cooked without the involvement of pottery or that the herd composition did not include enough pigs for the random sampling in this study to detect them.

When looking at the characteristics of the vessel which used to contain dairy products, the wall thickness of sample CVP5 suggests significant quantities of milk could have been processed at once. If this premise is accepted, it seems highly unlikely that a single goat or sheep would have been able to produce enough milk to fill the analysed vessel. Therefore, it could be speculated that the vase might have contained either mixtures of different milks from different animals or that possibly animals with a higher milk production (like cows) could have been milked too. In consequence, the organic residues recovered from the pothserds in El Cavet could be the result of managing a herd whose composition could be similar to that found in other contemporary sites (sheep/goat and cattle with a minor amount of pigs and wild species). Even in the case of sample CVP2, which was located in a feature from a possible more recent period in the Early Neolithic (2016.1), also presents a residue associated with the consumption of ruminant adipose fats.

In terms of the possible cooking strategies practiced in the site, although no molecules supporting vessel temperatures above 250ºC during use were detected, the cooking of animal fats remains one of the likeliest explanations for the presence of said products in the pottery's clay pores.

6.2 Cova de la Font Major

6.2.1 Introduction

The “Font Major” cave is at l’Espluga de Francolí, a town in the Francolí valley which connects the Tarragona coastal plain with the inland. This is a natural communication path which encloses really long karstic systems (3950m of East to West galleries) is 400 meters above the sea level and roughly 40km away from the coastline (Oms 2014).

The cave’s location implies that human societies used it throughout history. They were a powder keg during the Civil War and hikers and excursionists repeatedly visited the cavities afterwards until the town council closed the entrance to prevent looting (Cebrià *et al.* 2014). Across time, several archaeological excavations with a wide variety of methodological expertise have revealed upper Palaeolithic, Early Neolithic, Bronze Age and Iron Age occupations (Cebrià *et al.* 2014). Studies on the excavation results helped renovate of the cave’s museistic discourse. In 2011, a project aiming to update the infrastructure incorporated a new excavation that recovered the cavities’ full morphology by removing the sediments preserved behind a modern wall (Cebrià *et al.* 2014).

6.2.2 Archaeological features relevant in this study

The 2011 excavation recovered a 2m stratigraphic sequence containing 8 archaeological layers. Although earlier work severely affected the uppermost phases, layers III and Ig, which were below the present-day circulation level, still contained a significant quantity of unaffected remains (Cebrià *et al.* 2014).

While layer III belonged to the river bed, and the materials found there had been weathered and transported in a secondary position, layer Ig was 20cm thick and presented over 1000 archaeological remains such as pottery, bones, charcoal and lithic industry. Analysis of an Ovicaprine molar yielded a 6310±40 14C date which fits in the Cardial Early Neolithic. The characteristic cardial decorations found in the potteries’ exterior surfaces also corroborate this absolute date (Oms 2014).

6.2.3 Archaeological materials

6.2.3.1 Pottery remains

Regarding pottery, previous excavations obtained an assemblage of 80 fragments temptatively assigned to the Early Neolithic. A lot of small vases found in older excavations and bigger vases from 2011 suggest different activities could have been performed in different parts of the cave. Furthermore, although cardial decorations were always present, the higher amount of incised surfaces suggested the researchers the older assemblage could also originate from an occupation more recent that level Ig (Oms 2014).

Alternatively, level Ig yielded 305 fragments which could have belonged to at least 32 different vases. These had been cooked under a mixture of reducing and oxidising atmospheres that result from firing pottery in open fires. The clays incorporated mainly limestone inclusions although the assemblage presented a significant amount of variability and the vessels' surfaces were either polished or smoothed and, in three cases, spatulated. The shapes detected were mainly ovoid/spherical middle sized vessels, big cylindrical vessels and small vessels with a neck. Finally the decorations were mainly cardial impressions with plain appliqués and other marginal techniques (Oms 2014).

6.2.3.2 Lithic tools

The lithic tools in this layer were made 90% in flint and mainly belonged to three different exploitation strategies including a production process focused in the preparation of blades. Two polished axes and several ochre fragments were also found (Cebrià *et al.* 2014).

6.2.3.3 Animal remains

Regarding animal bones, the assemblage presented 201 significantly weathered fragments. Cutting marks and thermal alterations were indicative that both wild and domestic bones participated in food production activities. In 56 bones, analyses indicated the presence of *Ovis aries/Capra hirucs* (NR=27, MNI=6), *Cervus elaphus* (NR=13, MNI=2), *Capra pyrenaica* (NR=5 MNI=2), *Lagomorpha* (NR=3, MNI=2), *Bos* sp. (NR=8, MNI=1) and *Sus* sp. (NR=1, MNI=1). In terms of the digestive system, 52 remains originated from ruminants (93%), 3 remains originated from cecotrophs (5%) and 1 remain belonged to non-ruminant animals

(2%). Only three seashells (*Columbela rustica*, *Cerastoderma edule* and *Glicymeridae*) were present in this layer, suggesting only a minor amounts of marine products arrived at the site (Cebrià *et al.* 2014).

6.2.3.4 Plant remains

Although no carpologic or anthracologic reports from the early neolithic layers were available, pollen studies also indicate the forest environment around the cave would have been characterised mainly by *Quercus* sp. and a minor presence of *Pinus* sp. and *Juniperus* sp. (Ballesteros 2009).

6.2.4 Sampling and analytical techniques

Therefore, the Font Major cave offers the opportunity to obtain data from a site in a strategic place controlling possible routes to the interior of the Iberian Peninsula. Performing organic residue analyses is necessary to better understand the role of pottery in the economic activities related with the exploitation of this environment. Moreover, the reported studies of animal remains make a comparison with organic residues possible. Given that this is not a coastal site, following this study's objectives, only a limited number of samples were analysed. The five selected sherds originated all from layer Ig, dated from the Cardial Early Neolithic (Table 43). All the samples belonged to rims from different vases submitted to similar firing conditions involving both oxidation and reduction reactions. While vases 7, 14 and 62 presented no decoration, vase 84 had a plain appliquée applied in the exterior surface. Vase 81 featured two plain appliqués, one in the exterior surface and one in the interior surface.

Lab ID	Sherd	Vase	Excavation	Depth	Part	Firing	Decoration	Weight (mg)
FM7	470	7	14/07/2011	393	Rim	Oxidising	No	101
FM14	301	14	12/07/2011	384	Rim	Mixed	No	103
FM62	375	62	13/07/2011	389	Rim	Mixed	No	103
FM81	2	81	07/07/2011	358	Rim	Mixed	Plain appliquée inside	102
FM84	278	84	12/07/2011	400	Rim	Mixed	Plain appliquée	101

Table 46: Analised samples from Cova de la Font Major.

After sampling underneath the vessel's inner surface, organic residue analyses were performed on 1g of pottery following a direct acidified-methanol extraction. Samples were submitted to GC, GC-MS and GC-C-IRMS at the ICTA laboratories.

6.2.5 Results and discussion

The five analysed samples in Cova de la Font Major have yielded significant amounts of residues (Table 44). Although the majority of the samples contained from 10 to 20μg·g^{-1}, one specific sample (FM81) reached 736.5μg·g^{-1}. This is one of the highest quantities of fat detected across the whole regional study and corresponds with the vessel including a plain appliquée in the interior surface. Except for this fact, there seems to be no correlation between the quantity of lipids and the pottery's firing conditions or the presence of decorations. Moreover, both the samples with the highest amount (FM81) and the lowest amount (FM10) presented plain appliqués.

ID	TLE	P/S	δ^{13}C18:0‰	δ^{13}C16:0‰	Δ^{13}C‰	Interpretation
FM7	11.4	1	-28.3	-28.1	0.25	Degraded non-ruminant adipose fat
FM14	12.1	1.2	-	-	-	Degraded animal fat with possible plant material
FM62	19.4	1.7	-28.6	-26.7	1.85	Degraded non-ruminant adipose fat
FM81	736.5	0.5	-	-	-	High quantity of degraded animal fat
FM84	10	4.3	-	-	-	Degraded archaeological residue.

Table 47: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in μg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio.

The majority of the samples contained similar profiles with mainly palmitic and stearic acids surrounded by a wide range of free saturated fatty acids (from C10:0 to C30:0), minor quantities of monounsaturated fatty acids (mainly palmitoleic and oleic acids) and minor quantities of alkanes. The palmitic to stearic acid ratio in these cases is around one (min 0.5, max 1.7). This profile, depleted of any plant or marine biomarkers, is nonetheless coherent with free fatty acids resulting from the hydrolysis of triacylglyerols from an animal origin. Nevertheless, sample FM84 presents a P/S value round 4, indicating significantly higher quantities of palmitic than stearic acid. Although researchers have suggested this value could be associated to an input from plant material (Copley *et al.* 2005e, Dunne *et al.* 2016), this interpretation cannot be backed by any other plant specific markers such as odd over even quantities of alkanes, plant epicuticular waxes or plant sterols. In light of this data, the nature of this residue remains unclear and has been classified as a degraded archaeological residue. Alternatively, sample FM14, although presenting a P/S ratio coherent with animal fats, yielded a range of linear fatty alcohols with 24, 26 and 28 carbon atoms accompanied by an increase in the amount of C24:0, C26:0 and C28:0 saturated fatty acids. Given this profile, it can not be ruled out that part of sample FM14's residue originated from wax esters (Matlova *et al.* 2017). The absence of C16:0 hydroxy fatty acids and the significantly low amounts of alkanes suggest this possible wax could not have originated from bees, but rather some plant. It is also possible that these alcohol and fatty acid duplets were originally present in the sample in free forms. The only way to clearly assess the origin of these compounds is to gain access to new a new sample from the same vase and extract the residues with a conventional dichloromethane:methanol extraction. Given that the hydrolysis of waxes could also increase the amount of palmitic and stearic acids recovered, a possible plant input would skew the result of an isotopic analysis. Therefore, this sample was not submitted to GC-C-IRMS.

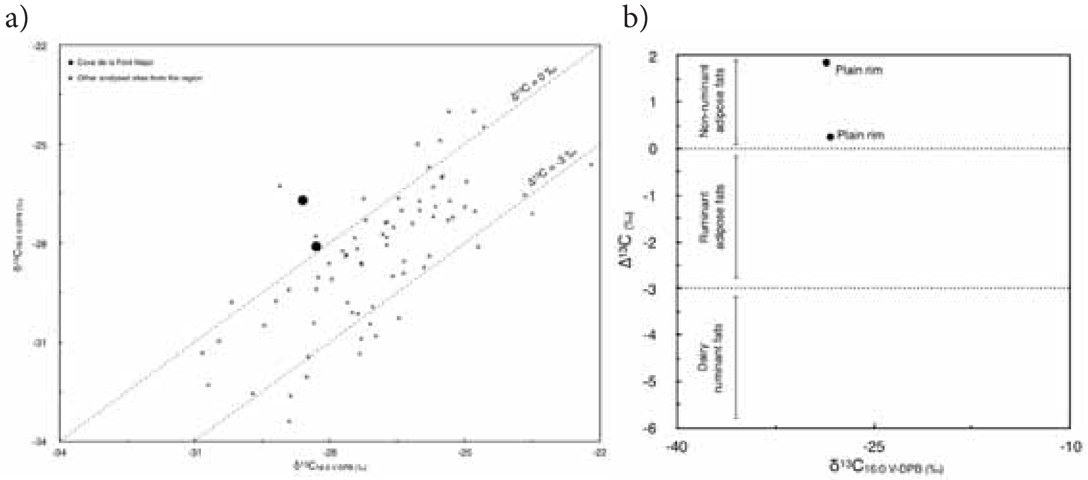


Figure 81: Results from the compound specific isotopic studies. a) plot presenting the δ¹³C:18:0 and δ¹³C:16:0 values b) plot presenting the Δ¹³C and δ¹³C:16:0 values.

Alternatively, samples FM7 and FM62 presented a significant amount of palmitic and stearic acids and a profile free of wax related compounds. Isotopic values for these two samples (see Figure 81) revealed the presence of Δ^{13}C values above 0, which has been associated with non-ruminant adipose fats. Moreover, the δ^{13}C16:0‰ and δ^{13}C18:0‰ absolute isotopic values indicate that these fats do not plot in the same area established for porcine adipose fats. Similarly from ruminant fats, which have been shown to vary across different climatic regions (Mukherjee *et al.* 2005), porcine adipose fats from Malta, the United Kingdom, Switzerland, the Middle East and Japan roughly plot around the same area with minor variations. Wild boars from Japan, Italy, Germany and the Middle East provide more depleted δ13C16:0 and δ13C18:0 values (Gregg and Slater 2010, Lucquin *et al.* 2018).

Figure 82 shows that samples from Cova de la Font Major plot outside the area for possible protein rich (domestic) porcine adipose tissues but inside the area for possible plant rich (wild) porcine adipose tissues. One of the many factors affecting isotopic values is the composition of the diet. In the case that other were minimal, it could be suggested that lipid residues from the two analysed samples in Cova de la Font Major originated from porcine fats feeding on mainly plant products. Other alternative explanations would be that these non-ruminant values originated from a range of mixtures or non-porcine species. Nevertheless, bone remains in the site only contain two non-ruminant species, one bone remain from Sus sp. and 3 bone remains from an undetermined Lagomorpha species. These mainly include rabbits, hares and other highly herbivorous species. Modern rabbit reference samples indicate adipose fatty acids are significantly more depleted than the values obtained for samples FM7 and FM62 (Taché and Craig 2015). Therefore, unless the sample originated from a non-ruminant species not detected in the animal bone record, the most probable option is that it belonged to a porcine fat.

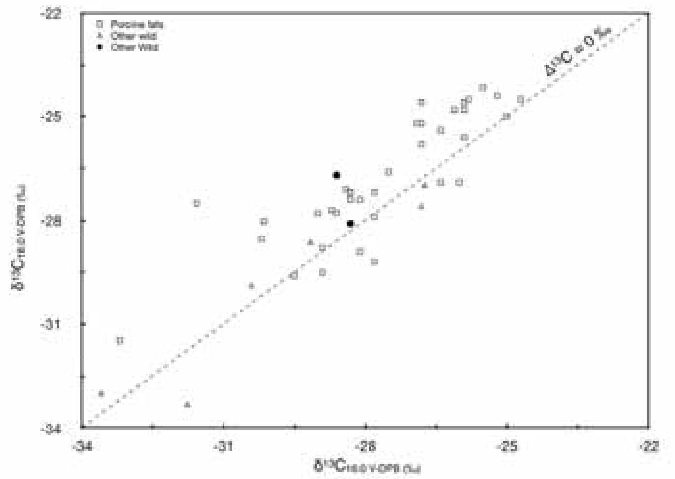


Figure 82: Comparison of $\delta^{13}C16{:}0‰$ and $\delta^{13}C18{:}0‰$ values from non-ruminant modern reference samples with samples FM7 and FM62 from Cova de la Font Major. See figure 57 for reference data.

Alternatively, given that lipids in pottery can be assumed to represent an accumulation of uses, the effects possible mixtures could have had in the obtained isotopic values should be taken into account. To this end, a bayesian mixing model including dairy, ruminant and non-ruminant end-members has been used to evaluate samples FM07 and FM62.

According to the model results (Figure 83), it is not likely the two vessels included any significant amounts of dairy products (median=11%). Although some quantity of isotopically enriched ruminant adipose fat should not be rejected (median=47%), the model does not reject that non-ruminant adipose fats could be one of the major sources (median=35%) as well. In the case of Cova de la Font Major, the model is taking into account the possibility that the presence of ruminant adipose fats mixed with non-ruminant adipose would still result in values similar to the ones obtained in the analysis.

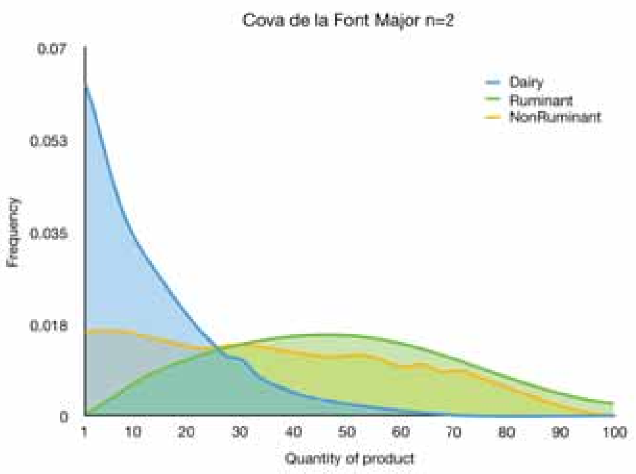


Figure 83: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from Cova de la Font Major.

It should also be noted that the sample with the highest lipid yields in this site is the one with an internal plain appliquée. More than a mere decoration, the possibility that this feature was intended for sustaining a lid should be taken into account. It is to be expected that those vessels more intensively used in cooking would be exposed to more food fats than other cases and, therefore, could easily present higher amounts of residues inside the clay matrix. In this sense, the existence of a feature which could be related with specific cooking practices (interior plain appliquée) is coherent with the significantly high amount of fats recovered. Moreover, given that the pottery sherd was sampled below the interior plain appliquée, lipids originating from that position would have most probably arrived there after several boiling episodes. Other cooking techniques such as stewing or frying must not be ruled out, as they would tend to concentrate cooking fats at the base of the vessel.

6.2.6 Conclusion

The analyses of 5 sherds from Cova de la Font Major suggest animal fats and possibly plant products were transformed inside the vessels. Isotopic analyses on two samples have been able to detect mainly non-ruminant adipose fats although the animal bone assemblage in the site is composed of 93% of ruminant species. These results might point at the possibility that significantly different cooking strategies were practiced in Cova de la Font Major, but it must be taken into account that the sample size is significantly low and, therefore, further studies could significantly change the interpretation of vessel use in the site. It must be stated, though, that despite the low amount of vessels studied, the vessels contained a varied range of products. Molecular markers suggesting but not proving the presence of plant products (possible hydrolysed waxes and high P/S ratios) were found accompanying the usual degraded animal fats. Despite the presence of marine seashells which could suggest some quantity of marine products could have been consumed in the site, no biomarkers for this type of products were found in the residues. In this case, said absence is not as significant as it could be in other archaeological sites closer to the sea. After all, Cova de la Font Major is 40 km away from the coastline, a stretch that would take more than 5 hours to cover by foot. In conclusion, the fact that for the inhabitants of this cave to directly access marine products they would have to walk for more than 10 hours, might present a limitation that could help explain the absence of marine biomarkers.

6.3 Cova del Vidre

6.3.1 Introduction

The Cova del Vidre is located at Roquetes, a town is in the Ports de Beseit mountain range. The cavity is 1100 meters above the sea level, approximately 30km from the sea, and has an eastern overture in the Lloret cliff, which. The cavity contains a triangular room and a rectangular gallery opening in the right side which were formed by a rift fold which has been cut by an encasing (Bergadà, 1995).

6.3.2 Archaeological features relevant in this study

This cave has been known to contain prehistoric remains since 1890. In 1915 Faura and Sanz excavated some trenches and recovered some archaeological materials, after that, Esteve Gálvez performed more excavations (Esteve 2000) and in the period between 1957 and 1960 Ignasi Cantarell systematically dug more trenches across the cave. The majority of the recovered materials were attributed to the Cardial early Neolihtic although non-cardial wares were also found within the assemblage. In 1992, new systematic excavations started by Josep Bosch and its team confirmed the stratigraphy detected by Cantarell and validated the superposition of Microlaminar Epipaleolithic and Cardial Early Neolithic without any posterior continuity. A test trench deep in the cave's interior did not detect Neolithic remains, but a wall or ceiling collapse with epipalaeolithic industry. Charcoal from this layer was dated from 10740±150 BP, which is well placed within the microlaminar period. Further geoarchaeological

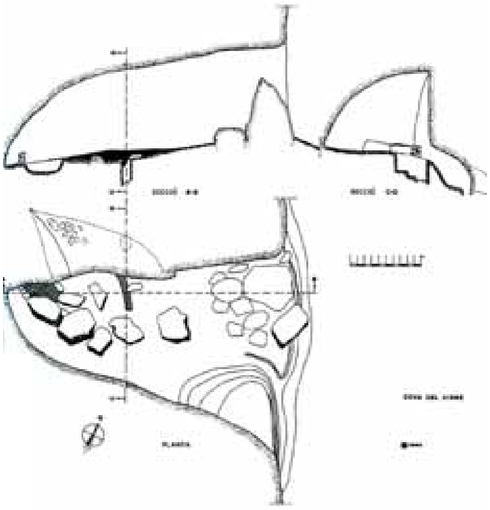


Figure 84: Plan from Cova del Vidre showing the excavated areas. Reproduced from Bosch 2016a.

analyses performed by Maria Mercè Berguedà (1995) fully characterised the four stratigraphic layers detected in the trench excavated closer to the cave's exterior (Figure 84).

Layer I: Composed of brown sandy silt 15cm deep, could be attributed to a modern deposition.

Layer II: Composed of brown sandy silt up to 53cm deep. Around 30cm down, an accumulation of charcoal remains in grey sandy silt was associated with a combustion structure. The material and the 14C date place this layer in the Cardial Early Neolithic. Several paleoambiental indicators point at tempered climatic conditions with a certain degree of humidity.

Layer III: Composed of light Brown sandy silt around 16cm deep with features suggesting oxidation and calcium carbonate concretions. It contained big blocks fallen from the cave's ceiling. The paleoenvironmental indicators suggested a cold and dry environment with undergoing icing and de-icing dynamics.

Layer IV: Composed of brown sandy silt more than 84cm deep. It was created in a cold humid environment (Berguedà 1998b). Charcoal from the combustion structure placed this layer in the Geometric phase.

The date of the epipaleolithic layers detected in Cova del Vidre by Cantarell and confirmed by Bosch *et al.*'s excavations was based on the presence of scarpers, bladelets and notched points which are found in other Microlaminar Epipalaeolithic sites (Bosch 2016a). The composition of the lithic industry suggested the assemblage could be placed in the Sauveterrian phase, linking the microlaminar phase with the geometric phase (Bosch 1993). Furthermore charcoal from the combustion structure in layer IV dated from 7290±70 BP and phosphated ferruginous impregnations possibly originating from animal remains in the fireplace were interpreted as indicative of culinary activities practiced in the fireplace. The low quantity of bone remains therefore was used to suggest these could have been for meat smoking or skin tanning (Bergadà, 1998b:145).

Cantarell also detected other layers at Cova del Vidre which could belong to the Neolithic horizon. Bosch *et al.*'s excavation validated them and recovered a rich assemblage containing cardial and non-cardial impressed pottery, appliqués, nipples, a blade based lithic industry, some geometrics, a significant quantity of bone industry and ornamental pieces. These were similar to significant Early Neolithic sites in the region (Bosch *et al.* 1993). Charcoal from this layer was dated from 6180±90 BP (Bosch *et al.* 1993) and, more recently, two ovicaprine bones found near the Neolithic hearth were also dated from the 6181±35 BP and 6248±33 BP (Bosch 2016b).

The combustion structure in the Early Neolithic layer was 3cm deep and contained vegetal and animal components. Charcoal, ash and limestone crystals formed around 500 to 600°C (Berguedà 1998b); calcined bone and phytoliths associated with ovicaprine coprolites, and fractures on the rock produced by thermal shock suggest, after Wattez *et al.* (1992), this was a primary combustion structure which would have been able to reach significant temperatures although other indices pointed at a short combustion time.

6.3.3 Archaeological materials

6.3.3.1 Pottery remains

To date, around 800 pottery fragments from all excavations in the cave have been studied by Bosch (2005). Although the assemblage presented a high fragmentation pattern, sherd widths between 4 and 13mm (mainly between 6 to 8) and rim diameters between 10 and 42cm (mainly between 14 and 19) suggest vessels recovered in the cave were mainly middle sized. These would be semi-spheric and open bowls and, more infrequently, globular closed with straight neck jars. The pottery surfaces were smoothed with significantly few polished fragments and the sherd colours (more grey than orange) suggest firing techniques involved open fires with poor control of the amount of oxygen in the atmosphere. In terms of decorations, roughly one third of the fragments were decorated by mainly non-figurative incised, impressed and plastic techniques. Nevertheless, one piece presents a slightly anthropomorph composition, similar to decorations in other Neolithic sites. Possible interpretations from this motive include a mother giving birth, which would invoke fertility and a possible maternal divinity (Bosch 2016b).

6.3.3.2 Lithic remains

In more detail, the recovered lithic pieces in the Early Neolithic layers were mainly made on flint and in one case, hyaline quartz. Blades, flakes and bladelets with simple continuous retouch and denticulates, scrapers, perforators, retouched flakes, notched bladelets, an abrupt retouched trapezoid, double bevelled triangles and half-moon segments also double bevelled (Bosch 2016a). Furthermore, polished stone tools were associated with grinding activities (Bosch 2016a).

The analysis of use-wear on lithic pieces from Level I incorporated 142 remains, 34 of which contained clear traces of use. Grouped by work categories, Butchering (n=14 41.2%), Hide scraping (n=9 26.5%) and Plant-wood crafting (n=5 14.7%) would have been the most common activities in the site. Hunting (n=3 8.8%), hide cutting or boring (n=1 2.9%), and the processing of Hard animal materials (n=1 2.9%) and herbaceous plants (n=1 2.9%) would also be present but neither sickles (n=0) nor use-wear related to mineral materials (n=0) was detected. In consequence, it seems Cova del Vidre could have been characterised by the presence of artisanal activities associated with leather and bone working. Moreover, it could be interpreted as a logistic camp where hunting and the processing of raw materials might have been the main performed activities. Some use-wear related to bone and wood present could also perhaps be linked to hunting around the site (Gibaja *et al.* 2018a).

6.3.3.3 Animal remains

The Early Neolithic layers in Cova del Vidre contained roughly 2360 bone remains belonging to at least 10 taxa, 951 of which could be identified to the species level (40.2%). Around 75% of the remains were classified as domestic. Ovicaprids were 89.01% followed by small amounts of *Bos taurus* (7,1%) and *Sus domesticus* (3.8%). Amongst wild species, wild goats were the most common (53,5%) but there was also a significant amount of *Oryctolagus cuniculus* (21.5%). Other wild animals were *Cervus elaphus* (18.6%), *Sus scrofa* (4,8%), *Capraeolus capraeolus* (1.2%) and Vulpes vulpes (0.4%)(Saña *et al.* in press). According to the mentioned quantities, ruminants would have been 90.3% of the remains, non-ruminants would have been 4.2% and cecotrophs would have been 5.5%. The animal assemblage in Cova del Vidre has been considered to present relatively lower percentages of domestic remains compared with other Early Neolithic sites (Antolín *et al.* 2018).

More precisely, rabbits and wild goat would be significantly more abundant that other common wild species such as deer, although this phenomenon might be explained by the topography of the site (Antolín *et al.* 2018). The study of kill-of patterns through age estimation indicates that an elevated number of animals were sacrificed before 6 months and between 6 and 12 months of age. Such early sacrifice ages could be coherent with the slaughter of animals before weaning, so the milk produced by the mother can be kept for human consumption (Saña *et al.* in press). Beyond dairy products, cutting marks were detected in 8.7% of the bones and a significant amount of the remains presented signs of thermic alteration (26.8%), which would clearly suggest that at least part of the assemblage was destined to human consumption (Saña *et al.* in press). Herbivore coproliths and phytoliths in square I-13 suggest possible animal herding in some specific places in the cave. However, remains in square J-17 suggest the presence of carnivores or omnivores with a high animal protein diet (Berguedà, 1995). Finally, piercers, a spatula a hafting implement and a small spoon support that at least part of the bone assemblage was used as tools.

According to Bosch (2016a) the Neolithic layers contained some marine seashell remains, some of them linked to body ornamentation. Two perforated shells (one *Columbella rustica* and one *Glycymeris* sp.) were accompanied by one piece of dentalium, discoidal perforated beads and a bracelet fragment on a *Glycymeris* sp. Other ornaments were made in bone (a ring) and in limestone (a bracelet). No other marine remains were detected in the Cova del Vidre's Neolithic layers.

6.3.3.4 Plant remains

No published data on plant remains from Cova del Vidre beyond the phytolith studies performed by Bergadà (1995) and the hearth's charcoal could be located in the literature.

6.3.4 Sampling and analytical techniques

Six potsherds were selected for organic residue analysis to evaluate the presence of lipid residues trapped in the clay matrix and gain knowledge on the state of preservation and quality of samples originating from old excavations. Three samples belonged to the Esteve Gálvez collection deposited in the Montsià Museum and another three originated from the 1992 excavations (Table 45).

After drilling around 1g of pottery, lipids were extracted with the acidified methanol procedure. Compound identification was carried out via GC-MS at the Department of Scientific Research in the British Museum and compound specific isotopic analyses were performed at the ICTA facilities.

Lab ID	Sherd ID	Museum ID	Year	Context	Shape	ø	Firing	Decoration	Plastic deco.	Weight (mg)
CVid7	107	29092	1992	K16	Rim	32	Mixed	Incised	none	113
CVid8	28	29102	1992	J16-17 K16-17	Rim	12	Oxidised	none	none	104
CVid9	117	29108	1992	J16-17 K16-17	Wall	-	Mixed	none	none	111
CVid10	44	1086	1960	Neolithic layer	Wall	-	Mixed	none	Handle	111
CVid11	90	1061	1960	Neolithic layer	Rim	40	Mixed	Incised wall and impressed rim	appliquée	103
CVid12	none	1047	1960	Neolithic layer	Wall	-	Mixed	none	none	111

Table 48: Analised samples from Cova del Vidre. ø: Rim diameter.

6.3.5 Results and discussion

All analysed samples presented lipid quantities higher than the accepted limit (5µg·g-1)(Table 46). Nevertheless, sample 1047 contained a quantity significantly lower than the rest of the assemblage. Given that some detected molecules in the majority of the samples (and also this one) have origins which could be partially or completely associated with postdepositional phenomena (alkane series and phthalate plasticisers), out of precaution, this sample has been considered a negative result for the presence of residues informing on vessel use. In consequence, 83% of the analysed samples yielded significant data.

Lab id	Context	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
CVid7	107	89.68	0.90	-22.2	-25.6	-3.4	Degraded and heated dairy product
CVid8	28	17.90	0.81	-	-	-	Degraded animal fat
CVid9	117	56.89	0.93	-	-	-	Degraded animal fat
CVid10	44	65.46	0.57	-26.5	-30.3	-3.8	Degraded dairy product
CVid11	90	23.18	1.88	-	-	-	Degraded animal fat
CVid12	1047	5.67	1.05	-	-	-	Non significant residue

Table 49: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

The 5 clearly positive samples presented a profile where Palmitic and Stearic acids dominated the chromatograms along with other saturated (C12:0, C14:0, C15:0, C17:0, C19:0, C20:0, C21:0 and C22:0) and unsaturated (C16:1 and C18:1) free fatty acids without any evidences against the residue originating from the degradation of animal fat triacylglycerols by postdepositional effects such as hydrolysis and oxidation. Moreover, the absence of key plant biomarkers and the presence of a significant amount of stearic acid related to palmitic acid would be a strong evidence against any major input of plant fats. Nevertheless, the residue from sample 90 contained a higher abundance of palmitic than stearic acid and a significant relative amount of unsaturated moieties. These pattern is not incompatible with fats from vegetal organic matter and therefore, this possibility must not be completely ruled out. Nevertheless, the absence of any other markers of plant organic matter such as sitosterol, wax esters and di- and triterpenoids suggest other alternative options might be more likely.

In conclusion, the range of molecules deleted would not be incompatible with the degradation of animal fat triacylglucerols due to complete hydrolysis and post-depositional degradation.

Sample 107 (Figure 85) is one of the most interesting vessels in the studied assemblage from Cova del Vidre. This is a rim fragment decorated with a triangular motive and filled with lines performed by a dragged incision. It contained the highest quantity of lipids for this site (89.7μg·g^{-1}) and a fatty acid pattern that, as it has been stated above, would not be compatible with a plant origin and not incompatible with an animal origin. Several peaks presenting the characteristic mass spectra of long chain ketones (*m/z* 239 and 255 for

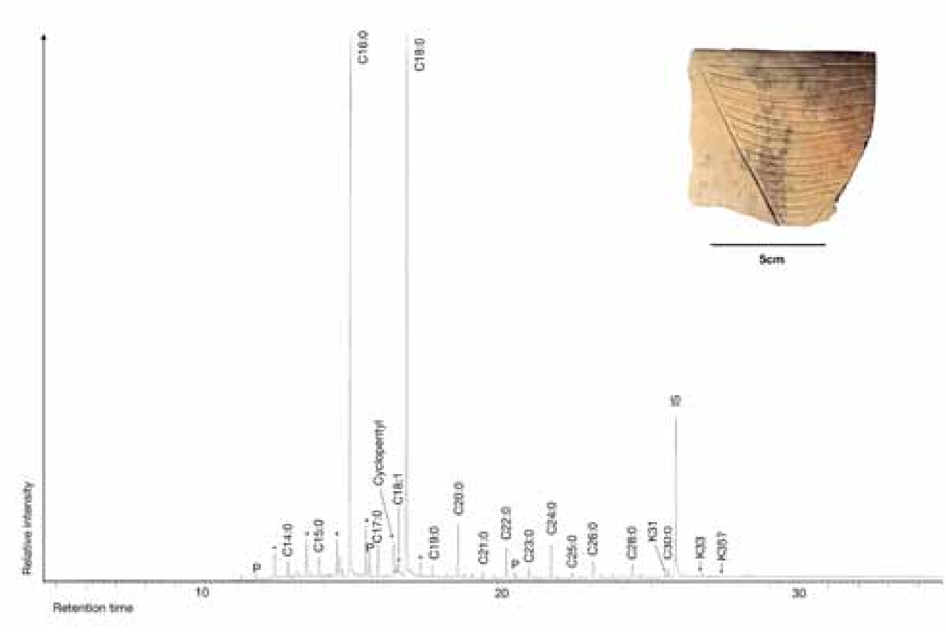


Figure 85: Chromatogram of sample 107 featuring the K31 and K33 and possible K35 long chain ketones and one peak identified as a cyclopentyl fatty acid.

16-hentriacontanone (31k), a combination of *m/z* 239, 255, 267 and 283 for 16-tritriacontanone (K33) and traces of *m/z* 267 and 283 pointing at the possible presence of 18-pentatriacontanone (35K) have been securely identified in the sample chromatogram. Although one of the possible origins for a series of long-chain ketones is a higher plant leaf wax, research has shown that specific long-chain ketones can be pyrolitically produced by condensation of fatty acids derived from fats absorbed in vessel walls.(Evershed *et al.* 1995a, Raven *et al.* 1997). It has been proposed that these reactions would include a free radical-induced dehydration and decarboxylation of fatty acid salts (specially calcium salts (CaO and CaCO3) at temperatures usually in excess of 350°C (Evershed *et al.* 1995a, Raven *et al.* 1997).

The fact that the carbon number range of the ketones in sample 107 is coherent with the condensed fatty acids and the position of the carbonyl group in the long chain ketones is either between 16 or 18 carbon atoms implicates the pyrolithic option must not be ruled out. Alternatively, in the presence of only minor amounts of very long chain fatty acids and the absence of alcohols, odd-chained alkanes (Jetter *et al.* 2006) and other ketones such as n-nonacosanone or nonacosan-15-one (Evershed *et al.* 1991), the possibility that the residue contained any significant amount of plant waxes should be considered minimum. Furthermore, other molecules also associated with the thermal alteration of fat are cyclic carboxylic acids (Hansel *et al.* 2004, Cramp and Evershed 2014). Their origin is considered to rise via alkali isomerisation of poly- and monounsaturated moieties followed by a 1,5 hydrogen shift, E/Z isomerisation, a possible extra 1,7 hydrogen shift and an intramolecular Diels-Alder (IMDA) reaction.

These results would have only been observed in experiments where samples had been subjected to temperatures higher than 200°C. Amongst the range of cyclic carboxylic acids generated, ω–(o-alkylphenyl) alkanoic acids have been the only ones so far reported in archaeological vessels. Given that their formation starts with alkali isomerisation they are expected to form after absorption of the precursor compounds in the clay matrix. Alternatively, non-aromatic five- and six- membered monocyclic fatty acids have been show to form after the formation of allylic radicals, 1,6 and 1,5 hydrogen shifts and cyclisation (Dobson *et al.* 1996a and b) in oleic acid isomers when heated (Dobson and Sebedio 1999). The existence of these saturated cyclopenyls (CFAM) in archaeological samples has been considered (Hansel *et al.* 2004, Cramp and Evershed 2014) but has not been reported to date. Peaks in Rt=16.386 and Rt=16.401 for sample 107 revealed a mass spectra coherent with the one reported after methylation of saturated cyclopenyl octadecanoic acid (Sebedio *et al.* 1989) with the distinctive *m/z* 220, 246 and 278 ions (see figure 36). The presence of the K31, K33 and K35 long-chain ketones and also saturated cyclopentyl octadecanoic acid indicates that it is difficult to defend that the recovered both saturated and unsaturated fatty acids had not been thermally altered. The detection of these molecules indicates that this vessel's rim would have been exposed to temperatures of at least 350ºC. Given that the formation of CFAMs is not dependent on the presence of an acid or base catalysing agent such as the pottery matrix, it should not be ruled out that the product had been heated before becoming a residue and, once inside the clay pores, further heating would have created other clay-dependent thermal alteration products (long-chain ketones).

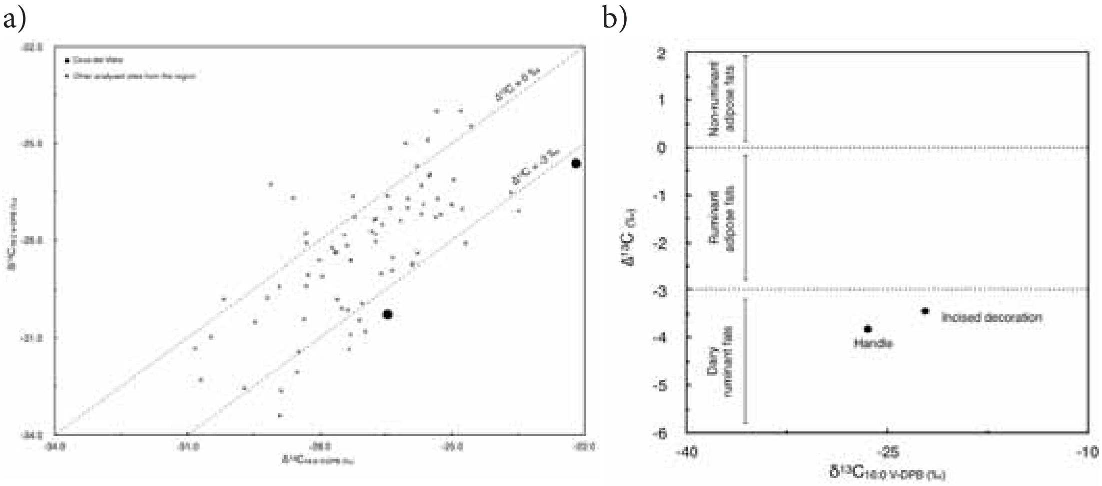


Figure 86: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

Due to the presence of UCMs and high background noise, only samples 44 and 107 could be submitted to compound specific isotopic analysis. Both results presented a Δ^{13}C value lower than -3‰, which has been consistently associated with dairy ruminant fats. Although wild ruminant species such as red deer have been shown to also present values below -3‰, their δ^{13}C absolute values were significantly more depleted (less than -27‰ for palmitic acid and less than -31‰ for stearic acid) than samples from Cova del Vidre (higher than -26‰ for palmitic acid and higher than -31‰ for stearic acid). These results could be coherent with the presence of a mainly ruminant dominated herd which, following the presented evidence, could have been herded inside the cave. The fact that the recovered lipids belonged to dairy products suggests animals could have been milked in the cave and the resulting product was transformed there. Cheesemaking or other processes removing excess lactose from milk do not need temperatures higher than 100°C. Therefore, the origin of the long chain ketones and cyclic acids might not be linked with the preparation of dairy foods, but their incorporation in everyday cooking practices as a finished product.

Although in Cova del Vidre compound specific isotopic analysis could only be practiced on two vessels, their signals could be considered an average of repeated cooking events possibly involving different products. To incorporate this source of variability into the interpretation, a bayesian mixing model including dairy, ruminant and non-ruminant end-members and excluding the possible presence of plant or marine end-members was applied to the data available for this site.

According to the model results, any significant amounts of both ruminant adipose (median=25%) and non-ruminant (median=15%) end members would not be solely capable of fully explaining the obtained isotopic values. Alternatively, it might seem that dairy products could have been almost exclusively used in vessels from Cova del Vidre (median=55%) but only further analytical work will be able to evaluate the extent and significance to herding management practices dairy products could have had.

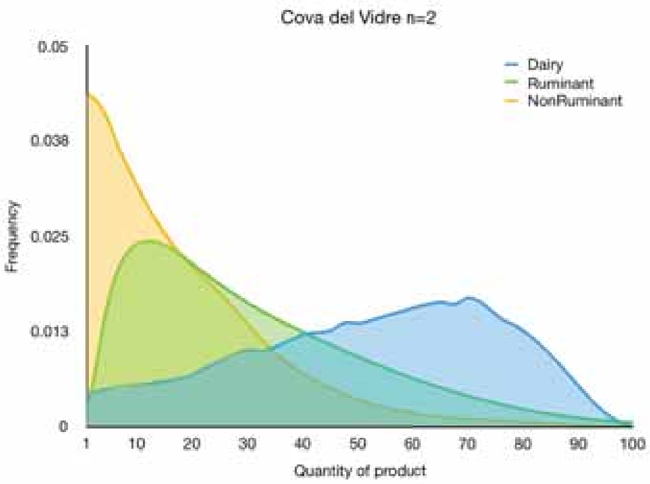


Figure 87: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from Cova del Vidre.

Across the site, the detected residues indicate a certain degree of variability in the types of products contained by pottery. As expected for Neolithic pottery in the mediterranean, the majority of the detected residues were in a high degradation state. Yet, the molecular study from these samples has been able to support a range of interpretations.

Isotopic measurements from samples 44 and 107 provide evidence from the use of milk and their possible transformation into dairy products, thus revealing the importance these might have had in this site. The long chain ketones and the cyclic fatty acids detected in sample 107 imply this vessel might reached high temperatures. Given the width and diameter of the pot, it seems that this would have been a significantly big vessel which could have been able to process significant quantities of food at once. This would be coherent with communal culinary practices. Although the sample size is small, the high percentage of positive results (83%) might indicate that the majority of the pottery located in Cova del Vidre would have been used in labor processes related to the storage and transformation of foodstuffs. Given that the best moment for lipid absorption into the clay matrix is when fats are in liquid state and that the majority of the detected residues had an animal origin it is difficult to defend that the vessels had been exclusively used for storage. Compared with residues from other archaeological sites, P/S ratios in Cova del Vidre all oscillate around 1. There is a complete absence of evidence for plant related products at least in the 6 analysed vessels. Furthermore, although some marine products reached the cave as seashells, no evidences for aquatic biomarkers were found in the residues. Therefore, had any transformation and consumption of such products taken place, the analysed vessels would have not played a recurrent role in it. During pre-Neolithic times, evidences suggest the cave was at least partially used to transform animal products. After the results of residue analyses are combined with evidences from the analysis of the hearths, it seems that, although the strategies used to access animal products were radically different, Cova del Vidre was still linked with this type of products.

6.3.5 Conclusion

Results from use-wear and animal remains studies suggest hunting and the incorporation of wild animals into the diet could have been more relevant than in other contemporary sites. Although the sample size in this case is small, so far organic residues point at the presence of activities focused on the management of the domestic range of activities also attested in the site by the remaining 75% domestic animal bone assemblage. At this stage it should not be ruled out that further analytical work could reveal the presence of residues coherent with the transformation of wild adipose fats using pottery, but, to date, there is no evidence for the existence of this activities yet.

6.4 Barranc d'en Fabra

6.4.1 Introduction

Barranc d'en Fabra is an open air site located in the right bank of the Ebro's lower course. It is near the modern delta and the city of Amposta in the Montsià region and roughly 4km away from the ancient seashore (Sornoza *et al.* 1998). The site is in the Carrova hill, an elevation 6 meters above the sea level which belongs to the elongation of the catalan littoral chain south from the Ebro and joins the Montsià and Godall mountain chains (Bosch *et al.* 1991)

This site was found during contract work when building the TV-3443 road, after previous reports (Baldellou 1971) indicated the presence of an Early Neolithic necropolis in the area. The main goal of the excavation was to locate said remains and, to this end, 18 trenches were dug around the area. One of them presented prehistoric remains, which motivated the extension of the excavation to 8m^2. This yielded even richer results but, before performing a fully open excavation, three long trenches were dug in order to assess the full extent and stratigraphic potential of the site. Budgetary issues and the construction of the road itself prevented the full study of the settlement and, by the end of the final fieldwork campaign, only 20% of a potential 1032m2 had been fully excavated (Bosch *et al.* 1995a). The site has, since then, been preserved below the modern road.

6.4.2 Archaeological features relevant in this study

As reported by the excavators, the site presented 4 main stratigraphic layers. The first was the modern sediment affected by present-day agricultural work. Below, layer 1 contained a significant amount of charcoal and sediment which was associated to a prehistoric event, the collapse of the settlement's buildings. Layer 2 presented the debris resulting from the domestic activities in the site and covered the remains of the structures built in stone. Finally, layer 3 belonged to a red and white sediment upon which the Neolithic human groups settled. Several postholes had been excavated in this last layer (Bosch *et al.* 1995a).

Although a full plan of the site is not available, infrared aerial photography detected the outlines of 10 round structures interpreted as 9 possible huts and 1 possible above-ground storage facility. Moreover, a set of adjacent smaller

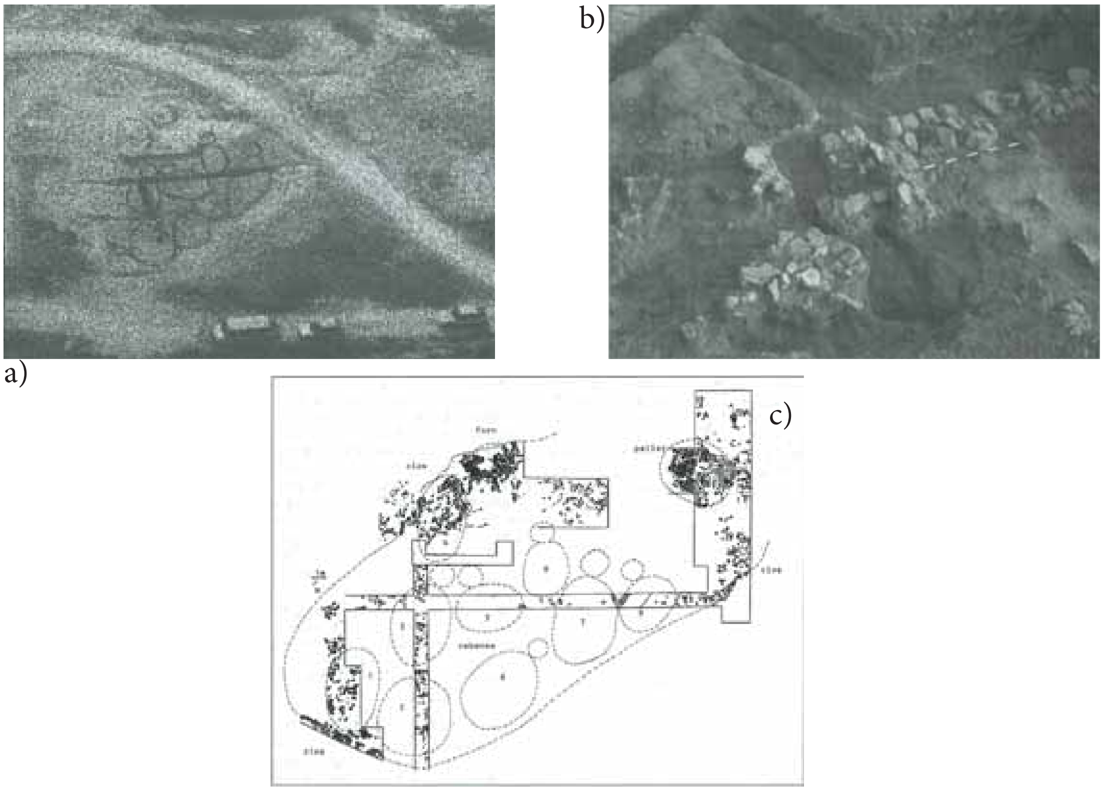


Figure 88: a) Infrarred aereal photography of the site. b) excavation picture showing the recovered walls. b) Site plan

features seemed to be in a position to support daily activities and a limiting wall encircled the whole group (Bosch *et al.* 1991). Two of the trenches partially crossed 6 out of the 9 possible huts and revealed mud-brick, stone walls and complementary postholes. The above-ground storage facility was laid out with flat stones around a central possible posthole and the excavation of the small annex features revealed significant quantities of ashes, charcoal and thermally altered clay, indicating these were probably combustion structures. Finally, the remains of a stone rampart were detected where the site limits could be excavated (the east, west and north)(Figure 88 a, b and c).

Following the excavators' interpretations, this was an open-air settlement showing a certain degree of spatial structuring. At least 6 of the 9 huts enjoyed and adjacent fireplace and all of them were grouped and limited by a long elliptical wall which, although existent, was probably more a representation of the inside versus the outside than a defensive feature (Bosch *et al.* 1995b). Charcoal obtained from one of the hut's postholes was ^{14}C dated from the 5880±110 BP. Further calibration at two sigmas using the IntCal13 curve identified the construction of the site at some point between the 5027 and the 4488 cal BC (Bosch 1993).

6.4.3 Archaeological materials

6.4.3.1 Pottery remains

The ceramic assemblage at Barranc d'en Fabra consist of 307 pottery very small fragments including 54 pieces characterised as rims, necks, bases and handles. Analyses of the sherds indicate that the assemblage would belong to at least 74 vessels. These would mainly be small or medium sized with diameters from 8 to 19 cm and wall thickness from 4 to 11mm. One outlier, a sherd 11mm thick and 52cm in diameter, has been considered to be a big vessel (Bosch *et al.* 1995a, Bosch 2005). Where shapes could be inferred, these suggested vases could be open or closed but never carinated. Decorations combined incised horizontal lines and dotted impressions, which are common in the final Early Neolithic and Epicardial Neolithic nearby sites (Bosch *et al.* 1995a, Bosch 2005). Other decorative features included straight appliqués in orthogonal disposition, third quarter moon shaped reliefs, which are highly infrequent and unparalleled in this region, and some combed surfaces (Bosch *et al.* 1995a, Bosch 2005). This range of stylistic designs is partially coherent with other assemblages, thus providing independent dating evidence complementing the obtained ^{14}C date (Bosch *et al.* 1995a, Bosch 2005). Nevertheless, it must be stated that the Epicardial designs at Barranc d'en Fabra contain specific motives which makes its ceramic assemblage unique in the context of the Early Neolithic in the northeast of the Iberian Peninsula.

6.4.3.2 Lithic tools

The range of activities practiced by the settlers at Barranc d'en Fabra included the manufacturing and use of lithic, macrolithic tools and pottery. Lithic tools were made on poor or medium quality flint and were mainly standardised blades which could have been hafted later (Bosch 1995a). Other types of lithic remains were nuclei, flakes and other undetermined pieces. For Barranc d'en Fabra, there were fewer retouched than non-retouched tools. Macrolithic remains mainly in sandstone were also found in the site but their morphology suggested that, instead of food processing, they were used for artisanal work.

6.4.3.3 Animal remains

Neither faunal remains which could shed light into the range of natural products consumed in the settlement were found in any of the site's features. The only direct evidence of animal exploitation is limited to a set of 34 remains of seashells 19 of which were necklace beads. Analysis of the remains revealed the presence of *Glycymeris violascens*, *Cerastoderma glaucum* and *Patella* sp., and evidences of thermal alteration and, in some cases, minimal use wear would point that part of the six complete shells and 13 fragments recovered would have been destined to human consumption (Bosch *et al.* 1995a).

6.4.3.4 Plant remains

Given the absence of charred seeds, plant data could only be obtained through the study of the recovered charcoals identified 12 taxa belonging to shrubs and one tree species, *Pinus halepensis* (Bosch 1995b). These suggest the vegetal landscape around Barranc d'en Fabra during Neolithic times was dominated by a bushy cover with small patches of pine woods.

The scarcity of direct paleoeconomic data made the study of the range of tools used in the site even more significant. Therefore, organic residue analysis on a selection of pottery sherds from the site provided an opportunity to gather relevant data on the possible animal and plant consumption practices.

6.4.4 Sampling and analytical techniques

Sampling for organic residue analyses performed on pottery from this site had to overcome the range of difficulties associated with small pottery fragments. No pieces below 10g were drilled in order to ensure the integrity of the sherd. In the case of three highly fragmented vessels, single and redundant wall sherds weighing roughly 3g were fully crushed to increase the sample size. Analyses were performed in a range of positions within the vessel including rims (n=4), walls (n=5) and a base. Although vessel 23 included a plain appliquée and vessel 32 had been incised and impressed, most samples, presented in table 47, were undecorated. Surfaces were mainly smoothed except for individuals 56 and 249, which were polished (Bosch 2005).

Lab id	Vase	Sherd	Surface	Colour	Shape	Decoration	Width	Diameter	Weight (mg)
BF1	53	249	Polished	Grey	Rim 1	None	9	13	108
BF2	15	281	Smoothed	Grey	Rim 3	None	7	13	96
BF3	23	260	Smoothed	Red	Rim 2	Plain appliquée	10	14	99
BF4	10	113	Smoothed	Red	Wall	None	11	19	199
BF5	2	217	Smoothed	Red	Wall	None	8	-	99
BF6	4	272	Smoothed	Red	Wall	None	6	-	101
BF7	34	118	Smoothed	Red	Base	None	11	-	112
BF8	32	56	Polished	Red	Rim 5	Incised and impressed	8	9.4	102
BF9	45	139	Smoothed	Grey	Wall	None	-	-	102
BF10	46	265	Smoothed	Grey	Wall	None	-	-	101

Table 50: Analysed samples from Barranc d'en Fabra.

The analytical techniques and methodologies used included obtaining roughly 1g of pottery and performing an acidified methanol extraction, GC-FID and GC-C-IRMS analyses. All work was carried out in the ICTA laboratories. The details on these techniques have been explained elsewhere (see section 3.3).

6.4.5 Results and discussion

After extraction, lipids were detected in all 10 samples. In the case of samples BF8 and BF10, the amount recovered was lower than the stablished threshold for possible soil contamination and, therefore, were considered to contain a non-significant residue. This implies that, for the case of Barranc d'en Fabra, the acidified methanol extraction was capable to recover significant residues in 80% of the analysed assemblage. In one case, the residue was accompanied by a set of not well resolved molecules. This phenomenon, known as an unresolved complex mixture (UCM), was minimal but significant enough to skew possible compound-specific isotopic analyses and, therefore, had to be discarded from further study. Other contaminations detected in samples from this site are the well known phthalic acids, which originate from plastic bags, but could be resolved and separated under chromatography.

ID	TLE	P/S	δ^{13}C16‰	δ^{13}C18‰	Δ^{13}C‰	Interpretation
BF1	15	1.9	-25.65	-26.89	-1.24	Degraded Ruminant adipose fat
BF2	49.6	1.3	-	-	-	Degraded animal fat
BF3	6	1.4	-26.47	-26.63	-0.16	Degraded Ruminant adipose fat
BF4	115.3	4	-	-	-	Possible plant oil
BF5	47.5	3.1	-	-	-	Possible plant oil
BF6	37.3	1.7	-27.29	-28.65	-1.36	Degraded Ruminant adipose fat
BF7	77.6	2	-26.72	-27.35	-0.6	Degraded Ruminant adipose fat
BF8	2.3	4.9	-	-	-	Non-significant residue
BF9	6	1.5	-	-	-	Degraded animal fat
BF10	4	2.2	-	-	-	Non-significant residue

Table 51: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

The lipid yields for Barranc d'en Fabra show a range of values from 2.3$\mu g \cdot g^{-1}$ (BF8) to 115$\mu g \cdot g^{-1}$ (BF4)(Table 48). When trying to explain the possible amounts recovered it seems that nor the vessel part (wall, rim, base)(Student's T = 0.479, p = 0.64), nor the colour (Student's T = 1.2281, p = 0.25) neither the wall thickness (Pearson's correlation coefficient r = 0.443 p = 0.27) correlate with the total lipid extract (TLE). It has to be point out, though, that the thickest sherds (11mm) were the ones to provide the most lipids (BF7 and BF4), but the thinnest vessel (BF2) also contained significant amounts of lipid residues (49.6$\mu g \cdot g^{-1}$). Despite vessel diameters could not be recovered in all sherds, which made the same size smaller (n=5), there seems to be a weak correlation between these two parameters (Pearson's correlation coefficient r = 0.86 p = 0.06). The smallest diameter (BF8) is also the most depleted vessel while the highest diameter (BF4) contains the highest amount of fat and the remaining sherds seem to be placed in-between (see figure 89). Regarding decorations, the sample with the plain appliquée (BF3) contained only 6$\mu g \cdot g^{-1}$ and the vessel with incised and impressed

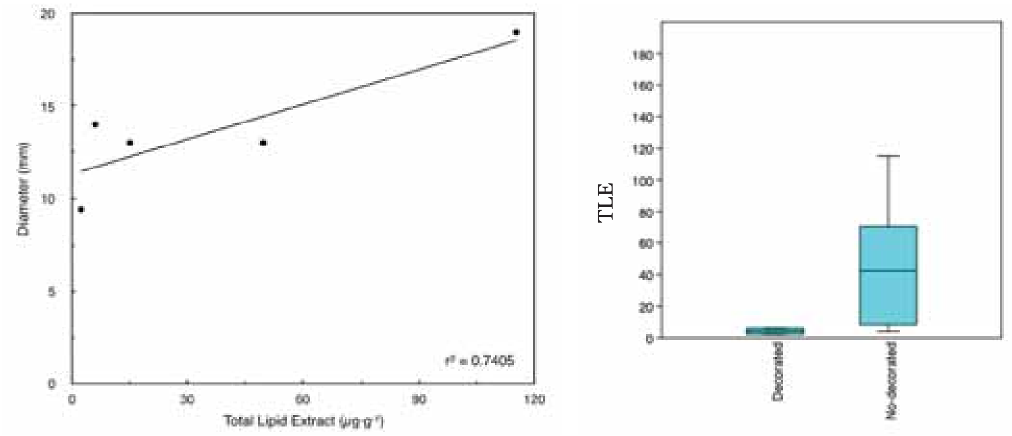


Figure 89: a) Comparison of vessel diameter and the Total Lipid Extract. b) Box plot comparing TLE and decoration.

decorations was also the one with the least amount of lipids. Nevertheless, the impact of the decoration on the possible amount of lipids can not be thoroughly assessed as more decorated individuals are needed to perform a Student's T test. When looking at the box plot, it could seem that, for this analysed assemblage, decorated vessels tended to contain fewer amounts of lipids than the rest.

Regarding residue identification, the majority of samples contained a series of free fatty acids. These ranged from C14:0 to C20:0 including palmitic acid and stearic acids, which were the two main moieties detected through gas chromatography. In some cases, significant amounts of C18:1 (oleic acid) were also recovered. The study of the Palmitic to Stearic acid (P/S) ratios could not rule out that samples BF5, BF4 and BF8 could have incorporated triacylglycerydes from a plant origin. This is interpreted given the presence of a much more significant quantity of palmitic acid than stearic acid (Copley *et al.* 2005e and Dunne *et al.* 2016). Nevertheless, the true nature of samples BF4 and BF5 remains unclear given the absence of other clear plant biomarkers such as odd-chanied alkane ranges or plant-specific sterols, waxes or resins. Although this is the case for 30% of the assemblage, the remaining 70% presents a similar amount of palmitic and stearic acids, a profile which would not be coherent with plant residues and has been more commonly associated with the degradation of triacylglicerols from animal fats. Compound specific isotopic analyses were performed in the four vessels with significant amounts degraded animal fats and the least amount of contamination possible. These were BF3, BF6, BF1 and BF7 (Figure 90).

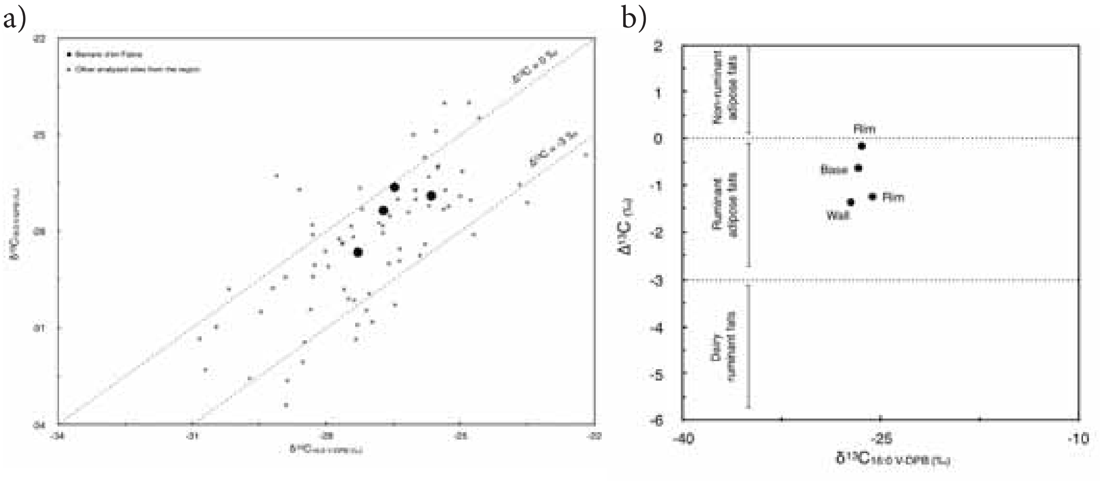


Figure 90: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

All four residues analysed by GC-C-IRMS presented $\Delta^{13}C$ values coherent with the presence of significant amounts of ruminant adipose fats. Signals for dairy and non-ruminant fats were absent although the small sample size for this case suggest caution should be taken. The presence of these two products should not be completely ruled out as values from BF7 could originate from a mixture of ruminant fats and a small amount of non-rumiant matter. Nevertheless, the present evidence only offers certainty that 100% of the analysed sherds contained a non minimal quantity of degraded ruminant adipose fats.

To further asses the extent of possible mixtures in pottery vessels from Barranc d'en Fabra, a bayesian mixing model was applied to compound specific isotopic values (Figure 91). Given these might result from an average of several vessel uses, the possibility that only one type of animal fat had been contained seems less likely than the alternative. In this case, dairy, ruminant and non-ruminant adipose fat reference values have been used as the end-members while other possible products such as plant or marine lipids were not incorporated due to the absence of relevant biomarkers.

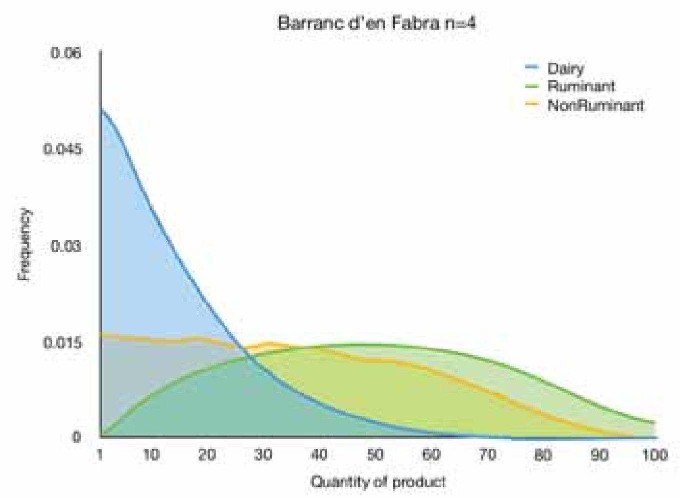


Figure 91: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from Barranc d'en Fabra.

The outcomes from the model seem to indicate that, overall, had dairy products been processed in the analysed vessels, quantities should be rather low (median=12%). Alternatively the model does not reject a possible higher presence of non-ruminant adipose fats (median=34%) although the probability ruminant adipose fats were the main product transformed seems to be higher (median=48%). In conclusion, organic residue analyses seem to reveal the use of these vessels as, at least, containers of ruminant and possibly also non-ruminant adipose fats.

Despite the absence of animal bones in Barranc d'en Fabra, organic residue analysis has been able to provide evidence for the transformation and consumption of products from an animal origin. Although no direct evidence from use-wear analyses is available, lithic tools could have been also used to manage animal products in the site. Furthermore, despite the study of seashell remains suggested they had been consumed, no biomarkers for marine products have been detected in the residues. This absence is significant given that the site is less than 5km away from the theorised Early Neolithic seashore (Sornoza *et al.* 1998). Unless further organic residue analyses provide evidence otherwise, this could suggest that seashell consumption did not involve processing this food though pottery or that specific marine lipids were either not well preserved or have not been found yet.

6.4.6 Conclusion

Given that the majority of the recovered residues belonged to animal fats, it is to be expected that they would have been solid at room temperature. It would have been therefore extremely difficult for any cold solid foodstuff to permeate through the clay matrix. Therefore, although no long chain ketones (Raven *et al.* 1997), ω-(o-alkilphenyl)alkanoic acids (Hansel *et al.* 2004) or other thermal alteration biomarkers were detected, it seems highly probable that the absence of some degree of heat, which would have been necessary to facilitate the incorporation of lipids into the pores, can not be rejected. This conclusion introduces the necessity to evaluate whether the analysed sherds could have been cooking vessels. It should be taken into account that although cooking pots would be in the best conditions to absorb the residues compared to storage vessels, tableware could also easily incorporate animal lipids as residues given that food would have been served hot and, therefore, lipids would still be in liquid form. To better clarify which options are more probable, vessel shapes and sizes must be taken into account. In the case of Barranc d'en Fabra, the high level of fragmentation prevents any detailed study of shape-use relations but results suggest the assemblage would have been composed by mainly medium to small sized vessels (Bosch 2005). This would point at the possibility that significantly small vessels had been used to cook minimal quantities of food. This interpretation could only be likely if vessels were used to prepare ingredients for recipes where the main source of food was being cooked following another method. Furthermore, data from other sites in the same period (see chapter on Reina Amàlia) suggest significant quantities of food were being cooked together.

Alternatively, the use of these vessels for illumination would facilitate the creation of molecules indicative of thermal alteration, but their absence in the vases from Barranc d'en Fabra suggests this was not the case. It may be more likely to theorise that these vessels could have been used as tableware, serving as a medium for the individual consumption of food, still within a group, or other specific uses involving the presence of animal fats.

6.5 El Molló

6.5.1 Introduction

El Molló is an open air site close to the modern village of Mora la Nova, roughly 30km away from the sea across the catalan prelitoral mountain range and around 55 meters above the sea level. It is placed on top of gravels, sands and clays originated from the alluvial fans and terraces from the Ebro river. After initial works by Espadaler (Bosch 2005), which excavated a test trench and recovered materials associated with the Neolithic and Bronze Age periods, the land was selected for the development of a new industrial area, which triggered preventive contract archaeological works. After several surface prospections and new test trenches, two wide excavations studied 11000m^2 but focused only on the areas directly affected by the development works. More recently, the bulk of the archaeological results have been integrated into a comprehensive study undertaken by the Autonomous University of Barcelona. Detailed reports on the remains from this site are forthcoming.

6.5.2 Archaeological features relevant in this study

The Molló archaeological site contained up to 71 negative structures, 10 of which were linked with Neolithic human occupations (Figure 92). The study of the refilling materials indicated that features 2 (E2) and 3 (E3) most probably dated from the Epicardial Early Neolithic (Piera 2010a, Piera *et al.* 2016). The first one (E2) had a circular plan measuring 90cm in diameter and in 15cm in depth. The second one (E3) was 30cm deep and its plan was ovated with a diameter also 90cm wide.

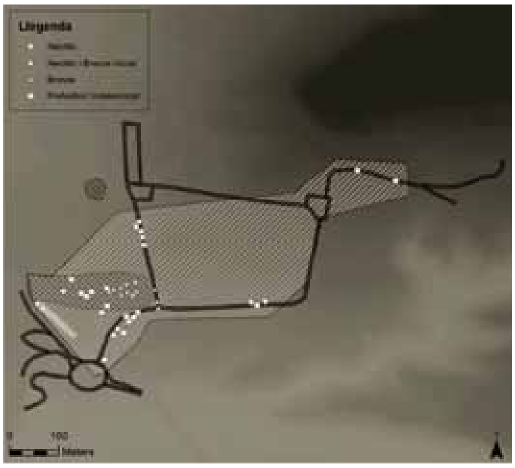


Figure 92: General plan of the archaeological structures found in El Molló. Reproduced from Piera *et al.* 2016

6.5.3 Archaeological materials

Pottery inside the structures was decorated with plain appliqués in horizontal and curved positions and motives were performed with impressed and incised techniques (Piera 2010a, Piera *et al.* 2016). Lithic tools and animal remains were also recovered in the structures' refillings, but the significantly high degree of fragmentation of the bone assemblage prevented their classification into species. Pending possible future work which may or may not provide new information, now it can only be initially stated that they belonged to middle sized mammals (Alcantara, pers. com.).

6.5.4 Sampling and analytical techniques

In the case of El Molló, organic residue analyses offers the opportunity to provide more information into the economic strategies practiced by the farmers and pastoralists inhabiting the Ebro's fluvial terraces in the Early Neolithic. To this end, 3 samples from the 1983 excavations located in the Tarragona Archaeological Museum and 4 samples from the 2010 fieldworks deposited in the Terres de l'Ebre Museum were selected for organic residue analysis. Samples from Espadaler's work belonged to layer G, which has been linked to the Neolithic occupations, and sherds from 2010 were found in features 2 (n=2) and 3 (n=2). As presented in Table 49, 4 samples were walls, 3 belonged to rims and two pieces were decorated with incised appliqués. The firing conditions were similar to the common practice for this period, where the entrance of oxygen was not controlled enough to ensure fully reduced or fully oxidised pieces.

ID	Year	Context	Layer	ID	Shape	Firing	Decoration	Weight (mg)
M1	2008	E3	30	-	Wall	Mixed	None	101
M2	2008	E2	20	3	Wall	Oxidised	Incised appliquée	108
M3	2008	E2	20	7	Wall	Mixed	None	101
M4	2008	E3	31	3	Rim	Mixed	None	104
M5	1983	9D	G	173	Rim	Mixed	None	114
M6	1983	9E	G	55	Wall	Reduced	Incised appliquée	109
M7	1983	9D	G	35	Rim	Mixed	None	111

Table 52: Analysed pottery fragments from El Molló.

Roughly 1g of pottery was pulverised and submitted to an acidified methanol extraction. Results were analysed through GC and GC-C-IRMS at the ICTA laboratories.

6.5.5 Results and Discussion

According to the total lipid extract, all samples in the site presented significant amounts of archaeological fats (Table 50). It seems that sheds from the 1983 excavation contained significantly higher quantities of lipids than the 2008 potsherds. Although it could be assumed that long term storage would significantly increase residue degradation, this was not the case. Instead, samples M5 and M6 yielded two of the highest lipid quantities in this study. When taking vessel parts into account, the three analysed rims yielded higher fat quantities than walls, except for wall M6, which presents the highest TLE in the assemblage. Alternatively, it doesn't seem that neither the firing techniques nor the presence of decorations affected the lipid yields. This situation may suggest two possibilities. One one side, vessels recovered in 1983 were affected by only minor post-depositional effects, preserving significant amounts of original fats inside the clay matrix. On the other side, post-depositional effects might have been as significant in both 1983 and 2008 excavations, but Espadaler's vessels could have already contained significantly more residues. If that second option were confirmed, it should not be ruled out that vessels participated in different activities than the sherds recovered in the silos.

ID	TLE	P/S	δ^{13}C16:0‰	δ^{13}C18:0‰	Δ^{13}C‰	Interpretation
M1	25.5	1.1	-27.62	-28.36	-0.75	Degraded ruminant animal adipose fat
M2	27.0	1.2	-25.9	-28.74	-2.8	Degraded ruminant animal adipose fat
M3	30.2	1.2	-25.79	-28.39	-2.6	Degraded ruminant animal adipose fat
M4	67.3	1.4	-26.73	-28.05	-1.3	Degraded ruminant animal adipose fat
M5	649.3	2.4	-	-	-	Degraded archaeological fat
M6	711.6	0.8	-27.04	-29.93	-2.9	Degraded ruminant animal adipose fat
M7	330.3	1.1	-27.65	-28.38	-0.7	Degraded ruminant animal adipose fat

Table 53: Results from the lipid extractions and the GC-MS and GC-C-IRMS analyses. TLE: total lipid extract is expressed in $\mu g \cdot g^{-1}$. P/S: Palmitic acid and Stearic acid ratio.

Regardless of the amount of lipids preserved, the samples revealed really similar free fatty acid profiles, including a range of saturated carboxylic acids with between 14 and 20 carbon atoms in their chains. Stearic and palmitic acids were the main molecules in the chromatograms, but other monounsaturated moieties such as oleic acid and palmitoleic acid were also detected in minor amounts. In many cases, the presence of phthalate esters in the samples could be interpreted as modern contamination from the plastic bags where the sheds had been stored. Finally, in all cases except one, the palmitic/stearic acid (P/S) ratio suggested that the free fatty acid profile would not be coherent with the

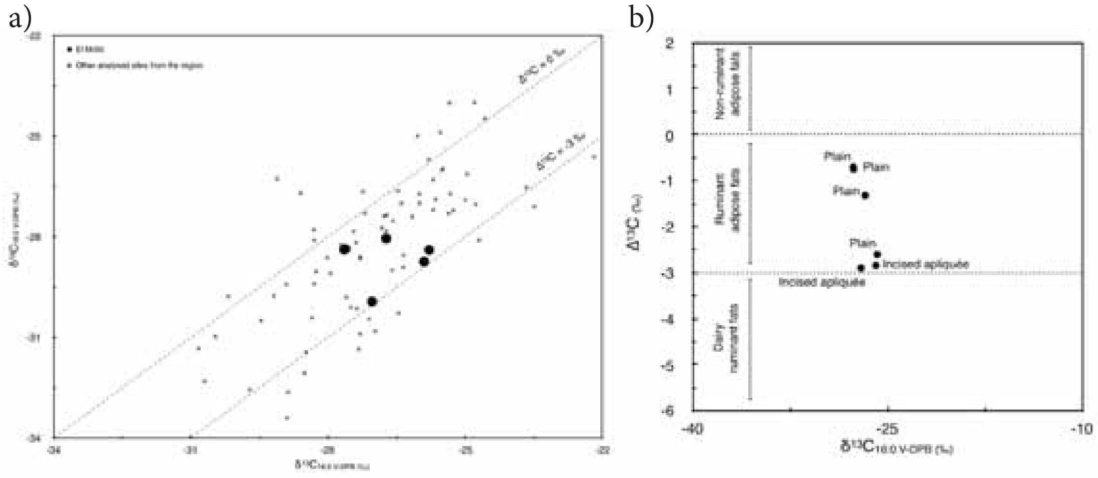


Figure 93: Results from the compound specific isotopic studies. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

degradation of plant triacylglycerides. Alternatively, the available data does not contradict the possibility that they had an animal origin. Nevertheless, for sample M5, a 2.4 P/S value would not be higher enough to point at the presence of residues from a plant origin alone, yet it might be high enough to suggest that the original residue could have presented a different composition or underwent a different degradation process than the other analysed samples. For this reason, sample M5 was excluded from further study via GC-C-IRMS.

Consequently, compound specific isotopic analysis was performed on samples M1, M2, M3, M4, M6 and M7. Results indicate that all samples fall within the boundaries for modern reference ruminant adipose fats. Nevertheless, it seems that, although all sherds present similar δ^{13}C absolute values, the Δ^{13}C index places them in two different groups. The first, samples M1, M4 and M7, plots around -1‰, and the second includes samples (M3, M3 and M6) with values significantly lower than -2‰. Given that -3‰ is the boundary accepted for a

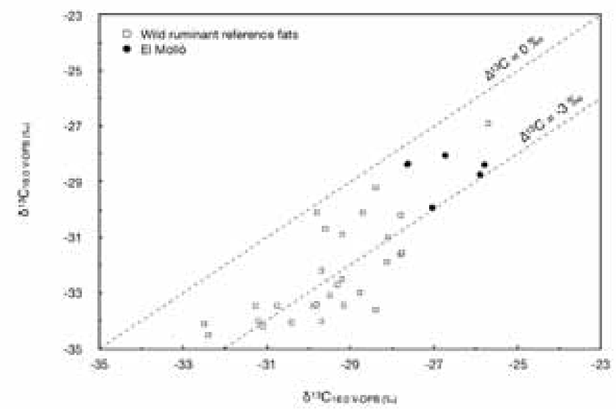


Figure 94: Comparison of δ^{13}C16:0‰ and δ^{13}C18:0‰ values from modern wild ruminant reference samples with samples from El Molló. See Craig *et al.* 2012 for reference data.

significant input of dairy (Spiteri *et al.* 2016) and 0 marks the threshold to differentiate between ruminant and non-rumiant animal fats (Spiteri *et al.* 2016), it should not be ruled out that one of the two groups contained minor amounts of another input different from pure ruminant adipose fats, or that the ruminant fats contained in the analysed sherds were subjected to different conditions which caused the variation in the $\Delta^{13}C$ values. Despite animal bone data not being species specific, it seems highly unlikely that the detected fats belonged to wild ruminant animals such as roe deer or wild goat. Modern European reference fats for these species usually present more depleted isotopic values (see Figure 94)(Craig *et al.* 2012).

To better assess the possible mixtures of animal fats present in El Molló, bayesian mixing modelling has been applied to the isotopic data. Given the absence of clear biomarkers for plant and marine products, the considered end-members have been ruminant dairy and adipose fats, and non-ruminant fats. This approach can take into account that mixtures of dairy and non-ruminant fats could plot in the ruminant area. Consequently, the results, rather than provide a single estimate for the possible mixture, explore all options and attribute them a probability. The addition and normalisation of all the values from El Molló, thus, provides an estimate of the importance each mixed product could have had in the strategies followed by this particular group.

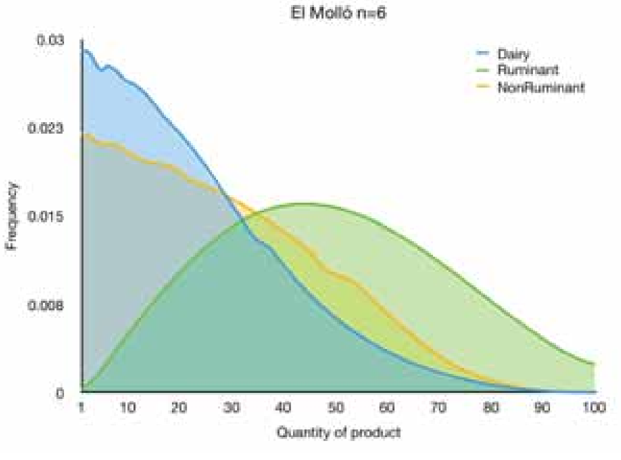


Figure 95: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in all analysed samples from El Molló.

According to the model (Figure 95), had dairy products been consumed in El Molló, the quantities present in the vessels would be significantly low (median=20%). Alternatively, mostly ruminant (median=47%) followed by non-ruminant (26%) products could have been more often prepared and mixed in the analysed pottery vessels. In consequence, it does not seem that, at least for the transformation of fatty animal products, any vessel could have been specialised.

6.5.6 Conclusion

After the presented evidence, mainly ruminant adipose fats contained in vessels could suggest pottery in El Molló was used to transform animal products originating from animal husbandry. Although it should not be ruled out that values from samples M1, M4 and M7 were coherent with a minimal input of non-ruminant fats, so far there is no direct evidence in the residues to support a significant amount of porcine cooking in pottery. If these species was ever consumed at El Molló, either residues supporting this hypothesis have not been found yet, or pigs were cooked following other techniques such as spit roasting. Whilst molecules supporting heating above 250°C were not detected, the fact that animal fats need some amount of heat to liquify and more easily permeate through the clay pores suggests that some of the analysed pots could have participated in the cooking of the detected animal products.

7 Cap de Creus

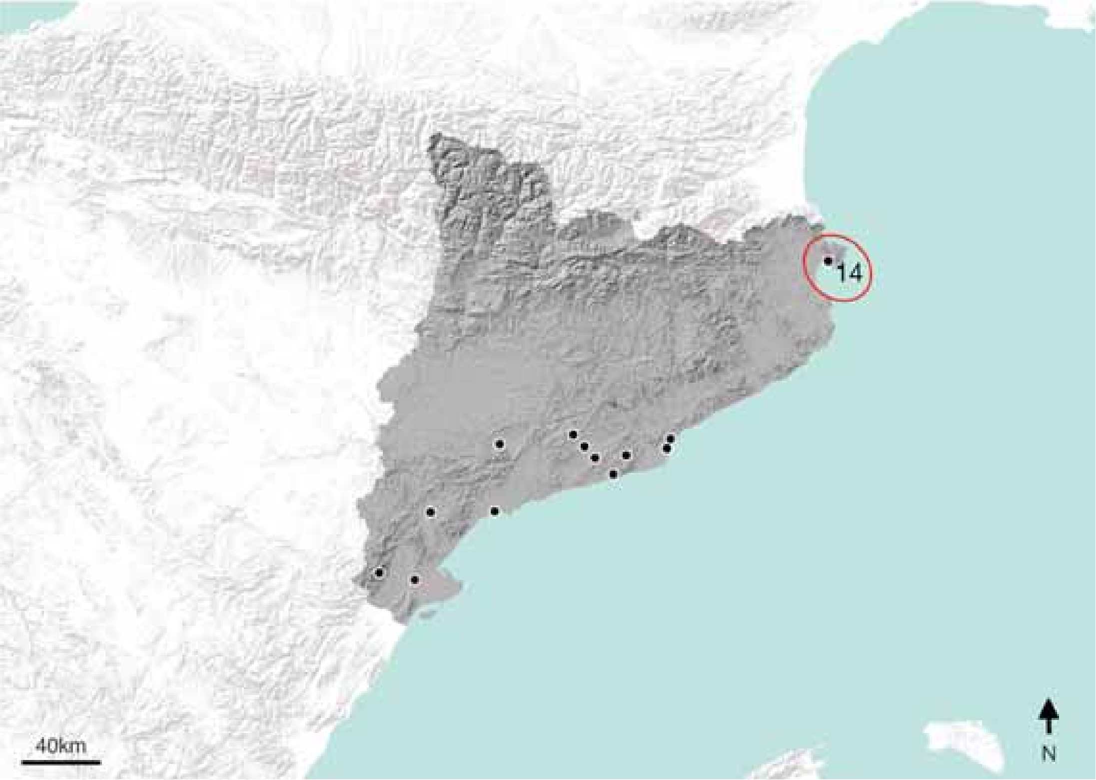


Figure 96: Map of the studied site in the Cap de Creus. 14: El Cau de les Guilles.

7.1 El Cau de les Guilles

7.1.1 Introduction

El Cau de les Guilles are two small cavities opened in the oriental Albera mountain chain, close to the modern city of Roses. These are less than 4km away from the beach and offer a view covering the majority of the ancient Empordà gulf, which has been gradually refilled through the Holocene (Blech and Marzoli 2005). The caves are located among a rich archaeological landscape including two megaliths (Dolmen del Cap de l'home i Dolmen Creu Cobertella) and a visigothic town (Puig Rom). Limited fieldwork halfway through the XXth century detected upper Palaeolithic (Magdalenian) and other earlier occupations. More recently, two campaigns between 2006 and 2007 excavated both the upper and the lower cavities using up to date fieldwork techniques (Soler i Serangeli 2008).

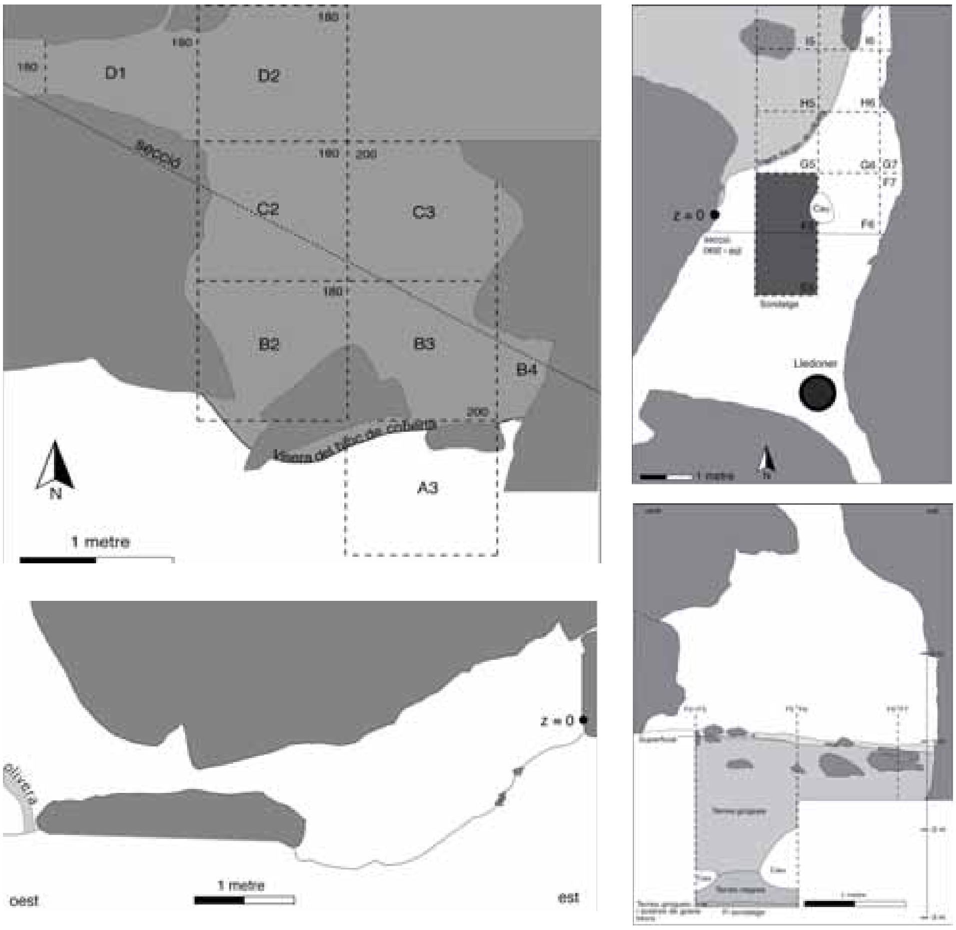


Figure 97: Cave plan and stratigraphy from Cau de les Guiles and Cau de les Guilles 2. Reproduced from Soler and Serangeli 2010.

7.1.2 Archaeological features relevant in this study

While the 2006 campaign focused on re-excavating layers from previous years, fieldwork in the 2007 reached lower non-affected layers only to certify that the cavities presented a high degree of bioturbation and post-depositional alteration (Figure 97). These phenomena prevented the recovery of any in situ stratigraphy but, after thorough work, a set of ceramic and lithic artefacts spanning from the Magdalenian to the Iron Age were recovered. Unfortunately, no faunal remains could be ascribed to any specific phase, thus preventing any thorough socioeconomic interpretations. Given the cavity's small dimensions and the presence of pottery remains the excavators did not rule out the possibility the site was used a storage space through the Neolithic (Soler and Serangeli 2010).

7.1.3 Archaeological materials

Out of the total 427 ceramic fragments recovered, the Early Neolithic occupation in these caves is represented by the existence of several sherds presenting unequivocal cardial decoration and a lithic triangle comparable to Early Neolithic sites in the Languedoc (Soler and Serangeli 2007, 2010). The main piece from this period is a fragment of a rim with a handle. This would have belonged to a hemispheric vessel 15cm in diameter decorated with seashell impressions both below, in the handle and following the rim. The decoration pattern in this piece also includes bands filled with horizontal incisions (Soler and Serangeli 2010). This specific technique which combines impressions and incisions has been extensively documented at La Draga (Bosch *et al.* 2000), which would be roughly 50km away (a distance which can be covered by foot in roughly 1 day).

7.1.4 Sampling and analytical techniques

Given the difficult archaeological context present in this site, organic residues offer a unique opportunity to gain new data on the socioeconomic practices which could have taken place in this point during the Early Neolithic. Studies on animal fat and/or plant and marine oils detected as pottery residues would help better understand the nature of the human presence in this cavity during the Early Neolithic. An assessment of the ceramic assemblage detected several small sherds whose decoration was coherent with an Early Neolithic chronology. Table 51 presents the 4 fragments selected for analysis.

ID	Site	Cut	Square	ID	Shape	Decoration	Chronology	Sample weight (mg)
1	CGUI 2	D2	BD	35	Wall	Incised appliquée	possible Early Neolithic	99
2	CGUI 2	B3	CB	30	Inflexion	Cardial impression	Cardial Early Neolithic	100.4
3	CGUI	H5	EH	23	Wall	Incised, garland motive	Early Neolithic.	99.2
7	CGUI	D2	BD	32	Wall	Cardial	Cardial Early Neolithic	42.3

Table 54: Analysed pottery fragments from El Cau de les Guilles (CGUI) and El cau de les Guilles 2 (CGUI 2).

Sample 1 is a body sherd presenting and incised appliquée. This is a common decorative technique in the Early Neolithic although it can also be found in other more recent periods (Manen *et al.* 2002). The potsherd originates from the lower cavity (Figure 98).

Sample 2 could possibly be part of a neck but the sherd's reduced dimensions prevent a definitive attribution. Two lines of Cardium edule imprints in the exterior face directly place this fragment in the Cardial Early Neolithic. This piece also originates from the lower chamber.

Sample 3 is a weathered and slightly bigger fragment whose decoration has been partially eroded. Although the decorative technique used can only be tentatively associated with the use of *Cardium edule*, the decoration's structure presents a set of semicircular concentric incisions limited by a series of circular impressions following the same axis. These are coherent with the garland Early Neolithic motive. Moreover, the circular impressions could have been easily created by using the seashell's umbo, which reinforces the interpretation that this sherd could have most probably belonged to the Early Neolithic occupation.

Sample 4 is a well preserved small fragment with a decoration and clay type analogous to the main cardial piece recovered (which was not sampled in this study). Its attribution to the cardial Early Neolithic is, therefore, certain.

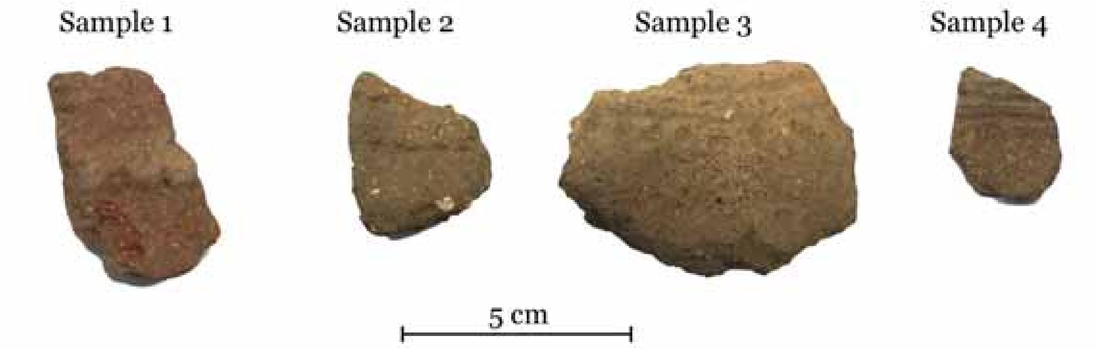


Figure 98: Exterior surfaces of samples 1, 2 3 and 4

Analyses were performed following the aforementioned techniques using the acidified methanol extraction, GC-FID and GC-C-IRMS. All analyses were performed in the ICTA laboratories.

7.1.5 Results and discussion

Out of the 4 samples analysed, 50% presents lipid quantities above the accepted minimal threshold (5μg·g^{-1})(see Table 52). Sherds 35 and 32 presented only trace quantities of organic matter. These minimal quantities are normally attributed to post-depositional effects and possible post-excavation contamination. Although the possibility that they previously contained an archaeological residue should not be fully ruled out, at the moment of the analysis, no significant amount would have remained to perform any significant interpretation.

ID	TLE	P/S	δ^{13}C18:0‰	δ^{13}C16:0‰	Δ^{13}C‰	Interpretation
35	2	1.9	-	-	-	Non significant residue
30	6.1	2.5	-	-	-	Possible degraded animal fat
23	630	0.2	-26.4	-28.5	-2.2	Degraded ruminant fat
32	3.45	-	-	-	-	Non significant residue

Table 55: Results from the lipid extractions and the GC-FID and GC-C-IRMS analyses. TLE: total lipid extract is expressed in μg·g^{-1}. P/S: Palmitic acid and Stearic acid ratio.

On the samples with enough lipids, sherd 30 presents a quantity close to the 5μg·g^{-1} limit, which suggests caution must be taken given that it is also possible that some of the lipids recovered originated from the surrounding soil. Moreover, the P/S ratio in this vessel (2.5) and the absence of plant specific biomarkers such as odd-chained alkanes, sitosterol, waxes and resin diterpenoids suggests the presence of any significant amount of plant matter might be rejected. Alternatively, the fatty acid profile is not fully incompatible with the degradation of animal fat triacylglyerides.

Finally, sample 23 presents a profile where Palmitic acid and Stearic acid dominate the chromatogram but other saturated fatty acids are also present (C14:0, C15:0, C17:0, C19:0, C20:0, C21:0, C22:9 and C23:0). Although no specific molecules associated with marine fats nor biomarkers from plant remains have been detected, this fatty acid profile is not incoherent with the hydrolysis of animal fat. Furthermore, experimental studies indicate that temperature animal fat absorption into the ceramic matrix given that these triacylglycerides would be solid at room temperature liquify. In this case and despite the fact that no biomarkers indicating high temperatures have been detected, the amount of fats recovered (630 µg·g^{-1}) is not against this vase being possibly associated with the preparation of food.

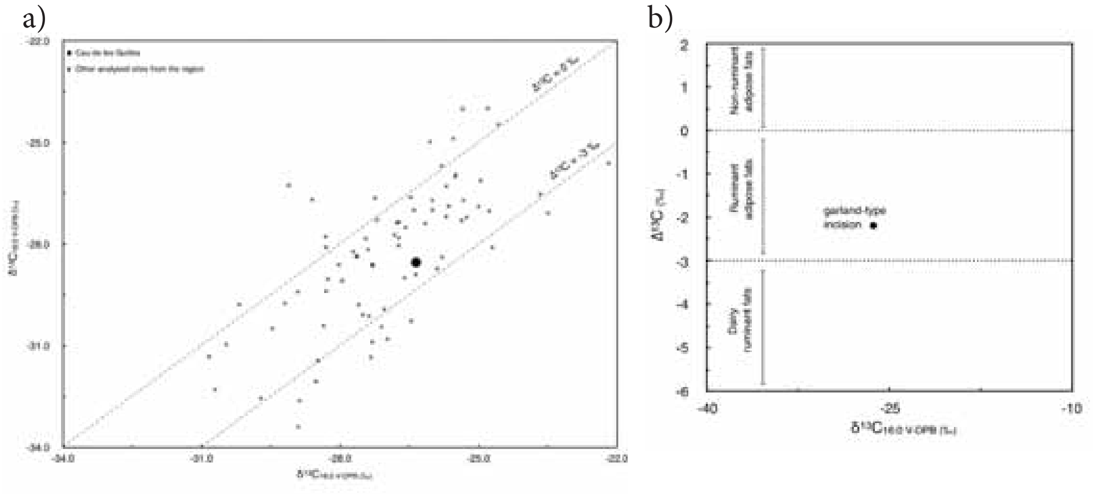


Figure 99: Results from the compound specific isotopic studies from sample 23 in El Cau de les Guilles. a) plot presenting the δ^{13}C:18:0 and δ^{13}C:16:0 values b) plot presenting the Δ^{13}C and δ^{13}C:16:0 values.

Compound specific isotopic analysis of the stearic and palmitic acids in sample 23 (Figure 99) obtained values not coherent with either subcutaneous fats from non-ruminant animals feeding on a mediterranean landscape (slightly enriched δ^{13}C values) and dairy products (Spiteri 2012). Further analysis of fats from modern wild animals have been shown to present significantly different δ^{13}C values (Craig *et al.* 2012, Luquin *et al.* 2018, Gregg *et al.* 2009, Taché and Craig 2015, Choy *et al.* 2016), which seems to indicate that the recovered fats at Cau de les Guilles would not be incompatible with the preparation and consumption of ruminant domestic species such as sheep, goat or cattle.

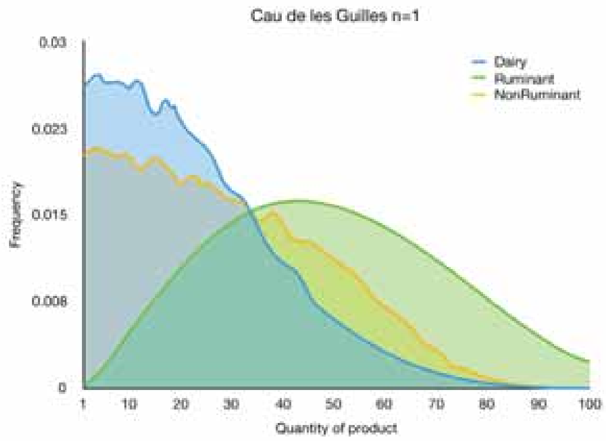


Figure 100: Modelled statistical distributions of possible dairy, ruminant and non-rumiant inputs in sample 23 from El Cau de les Guilles

Results from the bayesian mixing model including dairy, ruminant and non-ruminant end members (Figure 100) have been used to explore the presence of a mixture of products accumulated in the vessel through its lifetime. The resulting values do not rule out the presence of significant non-ruminant fats (median=26%) although ruminant adipose fats is the end member with the highest probability of being the major product preserved in the vessel (median=47%). Had dairy products been present, the model would not be compatible with high quantities of it (median=20%).

7.1.6 Conclusion

In conclusion, if it is assumed that the pottery located in the site was effectively used there, the recovered residues in pottery from el Cau de les Guilles suggest some domestic activities such as food preparation involving domestic ruminant adipose fats could have possibly taken place in the cave during the Cardial Neolithic.

8 Discussion

To summarise the aforementioned results, this study has analysed 114 pottery fragments from the Cardial and 76 pottery fragments from the Epicardial ceramic horizons in the northeast of the Iberian Peninsula. This total of 190 samples was selected from 14 single and multiple phased sites (11 from the 5500-5000 cal BC. and five from the 5000-4500 cal BC). For the Cardial horizon, in the Barcelona plain this included Caserna de Sant Pau, Berenguer de Palou. In the Garraf and Penedès area it included Guixeres de Vilobí, Cova de Sant Llorenç, La Serreta, Can Sadurní and Cova de la Guineu. In the southern area including the Ebro valley sites were El Cavet, Cova de la Font Major and Cova del Vidre. In the Epicardial horizon, the Barcelona plain included Carrer Reina Amàlia, Garraf and Penedès included Cova de Sant Llorenç and Can Sadurní and the southern area including the Ebro valley contained Barranc d'en Fabra and El Molló. In the Cap de Creus, Early Neolithic pottery sherds which could date from both the Cardial and Epicardial horizons were analysed from Cau de les Guilles (see Figures 18, 43, 47, 96 and Table 53).

Regions	**Sites**		**Samples**		**Significant residues**	
	Cardial	**Epicardial**	**Cardial**	**Epicardial**	**Cardial**	**Epicardial**
Barcelona Plain	2	1	26	52	22	48
Garraf and Penedès	5	2	51	7	36	7
South and Ebro valley	3	2	33	17	27	15
Cap de Creus	1		4		2	
Total*	10	5	110	76	85	71

Table 56: number of samples and positive results from the regions and chronologies incorporated in this study. *Does not include El Cau de les Guilles.

Rims and necks were selected when possible, but in some cases, due to the nature of the Early Neolithic ceramic remains, walls and bases were also sampled. The study included both incised, cardial decorated, undecorated fragments and pieces presenting appliqués and handles. The amounts of vessels containing a quantity of lipids attributable to an archaeological event were almost always superior to 50%. Only in Guixeres de Vilobí vessels with residues above the 5µg·g^{-1} threshold were lower than 45%. A total of 156 pottery sherds from the 14 Early Neolithic sites studied presented lipids whose archaeological origin could not be rejected (80%). Thanks to this data and previous work by Spiteri *et al.* (2016), the coastline between the Pyrenees and the Ebro mouth is, at this moment in time, one of the european regions with the highest amount of available data regarding organic residues in early neolithic pottery. The scope of the study is thus comparable to previous comprehensive research by Spiteri (2012) in the central mediterranean (69 residues from 10 sites), research by Evershed *et al.* (2008b) in the northern aegean coastline (158 residues from 9 sites), Ethier *et al.* 2017 and Cramp *et al.* 2019 papers in the Danube basin (109 residues from 12 sites) and Copley *et al.* (2005d) analysis of the southern British neolithic (189 samples from 6 sites).

From the 156 significant samples, 61 containing the profiles most coherent with free fatty acids resulting from the hydrolysis of terrestrial animal triacylglycerols were submitted to GC-C-IRMS. This was validated by the presence of cholesterol, P/S values coherent with degraded animal triacylglycerols and, in some cases, the detection of cholesterol.

The combination of the acquired data with the available knowledge of the archaeological context in which the pottery vessels were found has informed a set of discussion ideas. The initial one would be the exploration of possible geographic or chronologic differences in pottery use (section 8.1). One of the most substantial ones is the apparent dissociation between dairy and non-ruminant products in the studied sites (section 8.2). This phenomenon has motivated a deeper look into possible non-ruminant consumption practices (section 8.3) and the possible significance of high temperature markers as indicators of certain culinary techniques (section 8.4). The absence or minimal presence of other types of products such as plants (section 8.5) or marine organisms (section 8.6) has also been discussed in the light of complementary palaeodietary data. Finally, possibilities for the detection of fungal markers possibly related to fermentation have also been taken into account (section 8.7).

8.1 Geographic and chronologic differences informing trends in pottery use

Although variations across time and space might be detected, results from the 14 analysed sites indicate a common and general pattern. Most of the lipidic residues detected can be associated with degraded animal fats. When isotopic values are used to explore the range of mixtures which could have been contained in the vessels, ruminant adipose signals tend to dominate. This could be an artefact of the analysis given that the applied Bayesian model tends to overrepresent this type of product. Nevertheless, given that a significant amount of the measured $\Delta^{13}C$ values in the study are placed between -3‰ and 0‰, and that the majority of the zooarchaeological assemblages from the studied sites contain mostly ruminant bones, said dominance is feasable.

Within the Cardial period, a comparison of the animal fat types recovered between the Penedès and the South/Ebro valley shows only subtle differences which might be significantly altered with additional sampling. Nonetheless, it seems that Southern mixtures were slightly more enriched in ruminant adipose fats while the non-ruminant input seems to be stronger in the Penedès valley and Garraf massif region (Figure 101p). Dairy products in the south present a higher disparity of inputs with more mixtures being composed of either dairy inputs higher than 65% or lower than 30%. A specific comparison with the Barcelona plain might be complicated given that only one site from this territory has been studied. Nevertheless, across the three regions, the amount of mixtures which could have contained more than 50% non-ruminant adipose fats tends to decrease southwards (Figure 101m). While non-ruminant adipose fats are significant in Caserna de Sant Pau, La Serreta, Cova de la Guineu and Cova de la Font Major, no evidences of this product have been securely identified in El Cavet or Cova del Vidre.

Assessing possible chronological differences within the Garraf massif and Penedès valley region is not possible given that only five Epicardial residues from Can Sadurní and Cova de Sant Llorenç could be analysed (Figure 101k). Any comparison between the two periods would thus be unacceptably biased. Nevertheless, from a diachronic perspective, results from isotopic analyses in the Barcelona plain are significantly different between the Cardial and Epicardial periods (Figure 101a, d). Vessels from Caserna de Sant Pau tend to present higher amounts of non-ruminant products while evidences for a significant use of dairy products have only been detected in Carrer Reina Amàlia. Additionally, Epicardial samples seem to present slightly higher quantities of ruminant fats in their mixtures (Figure 101n). The Non-ruminant Cardial versus Dairy Epicardial association is completely reversed in the south (Figure 101o). Differences there are more subtle and could be affected by sample sizes, but they suggest that Epicardial vases could have incorporated higher amounts of non-ruminant adipose fats while dairy signals are stronger in the Cardial period. The only commonality between the Barcelona plain and the south would be the tendency to slightly increase the amount of ruminant adipose fats in the first half of the Vth millennium.

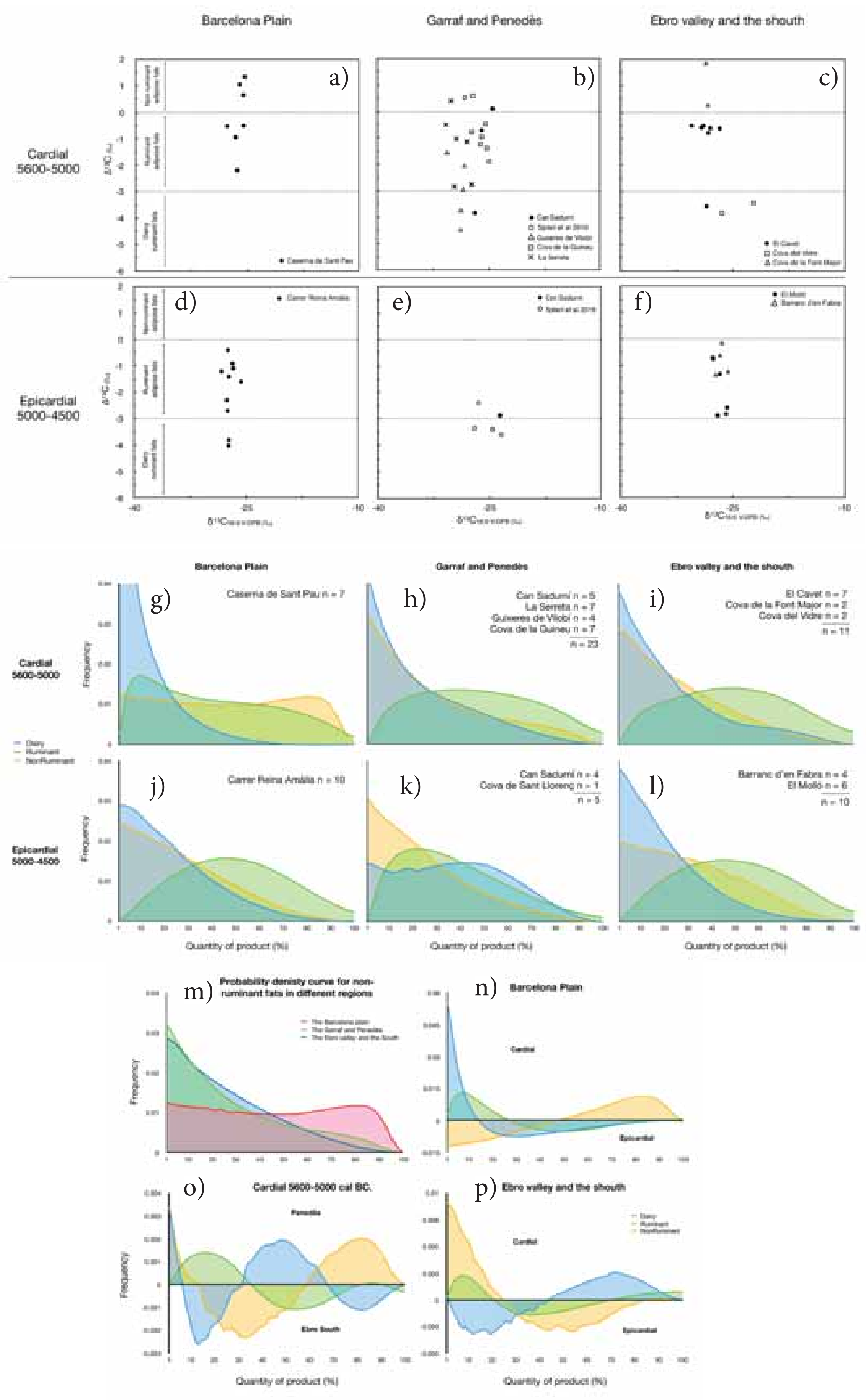


Figure 101: a-f) $\Delta^{13}C$ values from the studied regions and chronologies. g-l) bayesian statistical distribution of the dairy, ruminant and non-ruminant sources from the studied regions and chronologies. m-p) Differences between specific regions and chronological variations within specific regions.

It should be noted, though, that many of the chronological changes detected could be explained by the apparent disappearance of the north-east/south-west non-ruminant gradient in the Epicardial period. Therefore, it is of significant importance to study the distribution and prevalence of dairy ruminant and non-ruminant adipose residues in more depth given that they constitute the major changes across the studied space and time.

8.2 The mutual exclusion of dairy and non-ruminant fats

8.2.1 Study of the distribution in archaeological sites.

When taking into account all the ranges of compound specific isotopic values obtained in different archaeological sites, a possible mutual exclusion between isotopic signals coherent with dairy products and non-ruminant adipose fats seemed to emerge.

The first step to evaluate this possible association was to statistically validate that those sites with dairy signals did not contain non-ruminant adipose fats and vice versa. Almost all sites could be readily classified as containing one product or the other using the $\Delta^{13}C$ values except for Can Sadurní. In this site, the single sample presenting a value coherent with non-ruminant fats was problematic for two reasons; the first one was its PS ratio, placed at 3.8 and with presence of high heat biomarkers. The second one was a $\Delta^{13}C$ value at just 0.1, barely above the threshold for non-ruminant products. To increase the uncertainty, C18:0/C17:0br value did plot in an area incoherent with ruminant products and Can Sadurní was amongst the sites with the highest amount of samples containing isotopic values coherent with a major input of ruminant dairy products. In conclusion, although the presence of non-ruminant products in Can Sadurní should not be fully rejected, given the complexity in the interpretation of this case and for statistical purposes, Can Sadurní was considered to contain no significant amounts of non-ruminant fats within their analysed residues. Following these criteria, the following groups were formed:

1: Sites with dairy products and non-ruminant adipose fats: None. (N=0)

2: Sites with dairy products but without of non-ruminant adipose fats:

Guixeres de Vilobí, El Cavet, Can Sadurní, Cova del Vidre and Carrer Reina Amàlia (N=6).

3: Sites with no dairy products but presenting non-ruminant adipose fats: Caserna de Sant Pau, La Serreta, Cova de la Guineu, Cova de la Font Major. (N=4)

4: Sites with neither ruminant dairy products nor non-ruminant adipose fats: El Molló, Barranc d'en Fabra. (N=2)

These results for the whole 5500-4500 cal BC period were summarised in the following contingency table. When only the sites dated between 5500 and 5000 cal BC were taken into account, El Molló, Barranc d'en Fabra and Carrer Reina Amàlia were removed from the study. The data was studied using a χ^2 test assuming no association between ruminant dairy products and non-ruminant adipose fats as the null hypothesis (see appendix 4).

The results from the test do not completely agree with what can be intuitively detected by looking at the contingency tables. When all the sites in the study are taken into account, the statistical test suggests that the hypothesis of no association should not be fully rejected although the returned values are significantly close to the 0.05 threshold. Nevertheless, when looking only at sites belonging to the Cardial horizon, the dissociation between dairy and non-ruminant adipose fats becomes clearer.

This qualitative study, although presenting significant results, also contains several underlying limitations. Firstly, it is difficult to assess problematic cases such as Can Sadurní when a presence/absence decision must be taken. Had this site been considered to contain both non-ruminant and ruminant dairy fats, the χ^2 results would have probably been less indicative of an association. The second one is the fact that mixtures of products are not being taken into account. It should not be fully rejected that samples which plot closer but not within the expected values for ruminant dairy products could have contained minor amounts of it. The same case is valid for non-ruminant products but, moreover, particular mixtures of non-ruminant and dairy products could yield isotopic mixtures significantly similar to ruminant adipose fats, thus masking the presence of said products. To illustrate this effect, theoretical mixtures were calculated after Dudd et al's (1999) model for two (non-ruminant and ruminant) and three (non-ruminant, ruminant and dairy) sources using reference values reported by Fernandes *et al.* (2017). Three cases (no dairy, 10% dairy and 50% dairy) each containing 100 random ruminant and non-ruminant mixing values were generated using the RAND() function in excel (Figure 102). Results, as seen in figure x, indicate that the existence of a combination of ruminant and non-ruminant products with more than around 10% dairy might prevent the secure detection of non-ruminant fats.

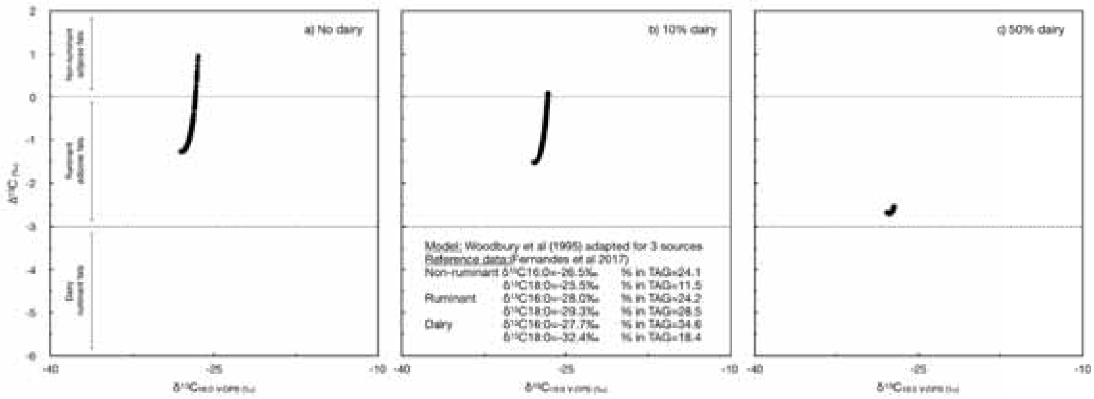
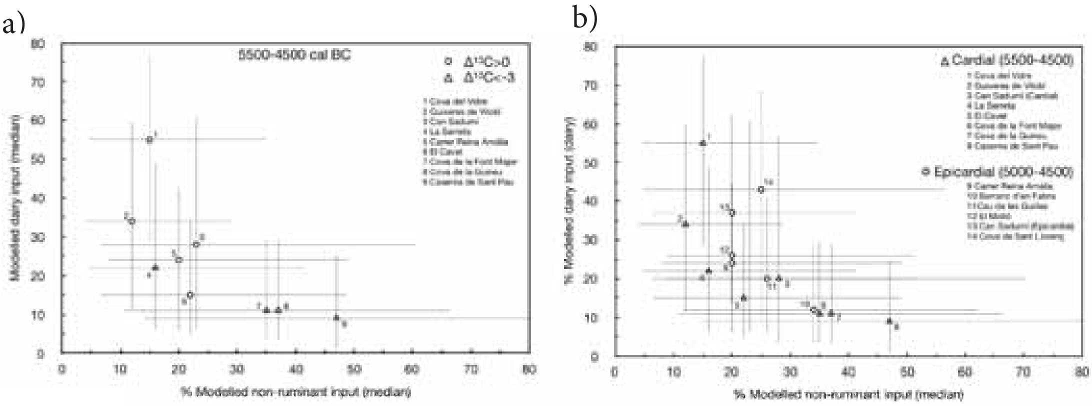


Figure 102: Simulated values for a random amount of ruminant and non-ruminant sources with an additional input of a) no dairy, b) 10% dairy and c) 50% dairy.

It is therefore necessary to construct a variable which takes possible mixtures into account and reflects the uncertainties associated with them. To this end, the results of the mixing model were studied. Given that its resulting distributions are not normal, the median, rather than the mean, could be considered as the best value representing the distribution. In consequence, the median values for the modelled amounts of non-ruminant and ruminant dairy fats in each site were studied to validate whether there existed an inverse correlation between ruminant dairy products and non-ruminant adipose fats.

Rather than treating each sample as the container of a product which is identified via biomolecular and isotopic biomarkers, the Bayesian mixing model (see section 3.5) assumes these values to result from a mixture of an unknown but significant amount of pottery uses. Later, all the available values from each site are averaged together, thus returning the expected relevance of a particular ingredient within a site. In this case, the quantity of samples averaged must be kept in mind as it might become a limitation when studying the correlation.

Although the statistical tests rejects the null hypothesis in all cases and correlation can be assumed, its strength was tested to further validate this effect. To this end, the r^2 value and the normality distribution of the residuals of a linear regression using the partial least squares algorithm were taken into account (see appendix 4).



and the percentage input of diary products (median) in a) sites containing samples with $\Delta^{13}C$ values either above 0‰ or below -3‰, and b) all sites incorporated in this study separated by chronological period.

Although the case including all sites presents a rather low r^2, removing the two sites with less than 3 studied samples improves the correlation. Residuals are normal in all cases, but the statistical result is significantly increased again when sites with less than 3 measurements are not taken into account. In essence, it seems that, the higher the amount of non-ruminant fats mixed in the studied vessels, the lower the amount of dairy products (Figure 103).

Nevertheless, this analysis contains a combination of two different historic periods (Cardial and Epicardial) which might be different from each other. Although the correlation seems to exist when combining them, they should also be analysed separately. Studying the possible correlation for the 5000-4500 cal BC range is not possible due to the low amount of sites studied, nevertheless, there are 8 cases available for the 5500-5000 age range and the results of the statistical tests suggests that the case of no correlation can still be rejected (see appendix 4).

Therefore, although it seems that the mutual exclusion of dairy and non-ruminant fats within residues of animal consumption in cardial pottery is significant, it is difficult to ascertain whether this practice disappeared or was maintained in the Epicardial.

8.2.2 Possible causes: caves and open air sites

A range of possible explanations for such phenomena might be searched by analysing whether the presence of dairy/absence of non-ruminants is affected by the characteristics of the archaeological site, or the composition of the herds available for cooking. To this end, the height of the site above the sea level (meters above the sea level, m.a.s.l), the sites' distance from the sea, the open air or cave nature of the site and the number of ovicaprine remains were taken into account. These variables were compared to a new indicator built as the difference between the expected median amount of non-ruminants and dairy products within each site (DairyMedian – NonRuminantMedian)(D-NR). Depending on the nature of the data compared and the expected normality of each variable, a set of statistic tests were performed (see appendix 4).

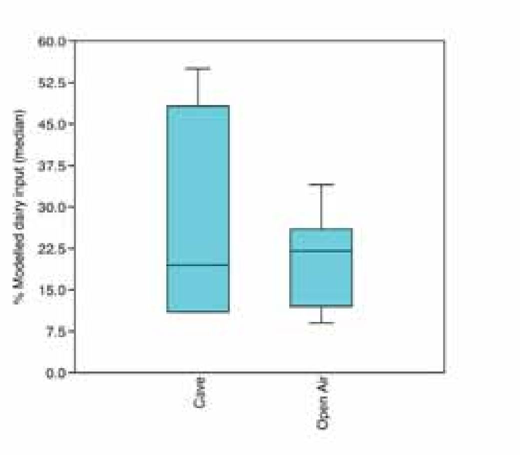


Figure 104: Box plot comparing the incidence of dairy products in caves and open-air sites within this study.

The first relevant result which must be taken into account is that there is no statistical evidence within the studied cases supporting the detection of substantial amounts of dairy residues solely on cave sites (Figure 104). This possibility had been previously contemplated by Spiteri *et al.* (2016) for the region of southern France and the Iberian Peninsula as an indicator of the possible existence of specialized seasonal herding sites. Caves and rock-shelters used as natural stalls could have been used as dairies, thus reflecting the significantly high amount of dairy products detected there. Nevertheless, residues presenting dairy isotopic signals from open air sites including Guixeres de Vilobí, El Cavet and Carrer Reina Amàlia suggest the existence of a more complex picture.

When the expected amount of dairy products mixed in the vessels is taken into account, a Student T test (N=11, T=0.68121, p=0.51) suggests that the null hypothesis that the mean amounts between the two groups are similar can not be rejected. It would initially seem that caves in the north-east of the Iberian Peninsula would not follow the pattern identified by Spiteri *et al.* 2016. Nevertheless, this does not take into account the available data within the western Mediterranean. Published studies for Early Neolithic caves and open air sites need to be taken into account to obtain a fuller picture that allows a discussion of the dairying cave hypothesis. To this end, a new comparison has been performed including 16 additional archaeological sites from the Adriatic to the Alboran Sea making up a total of 15 caves and 14 open air sites: Danilo and Pokrovnic (McClure *et al.* 2018), Mala Triglavca (Šoberl *et al.* 2014), Colle Santo Stefano (Salque *et al.* 2012), Font Juvenal, Grotte Gazel, Skorba, Grotta San Michele, Trani, Fondo Azzolini, Ciccotto and Nakovana cave (Spiteri *et al.* 2016), Pendimoun I, Grotte Lombard (Drieu 2017) Cova de l'Or (Martí *et al.* 2009) and layer IV in Cueva del Toro (Tarifa *et al.* 2019). When the percentage of lipid residues containing dairy signals is calculated and only sites with more than 3 available samples are taken into account, the resulting box-plot suggests that the association between dairy production and caves is still significant (Figure 105). This has been further confirmed by a Student's T Test (N=22, T=2.9409, p=0.008) which indicates that the null hypothesis that the means between the two groups are similar can be rejected. In consequence, when the results from this study are incorporated to the available corpus of analysis, the hypothesis laid out by Spiteri seems to be still valid.

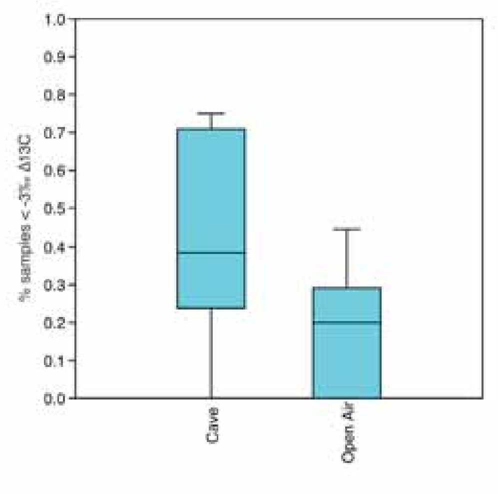


Figure 105: Box plot comparing the incidence of dairy products in caves and open-air sites from the western mediterranean.

Nevertheless, the neolithisation process in the western mediterranean has been shown to include two significant social developments, the Cardial and the

Impressa horizons (Bernabeu *et al.* 2009), which present different chronologic and geographic coordinates. When comparisons between caves and open air sites from these periods are performed, the two regions do not seem to be analogous (Figure 106). Nine sites with at least three reported samples (5 caves and 4 open airs)(Grotte Gazel, Can Sadurní, Cova de la Guineu, Cueva del Toro, Caserna de Sant Pau, La Serreta, Guixeres de Vilobí and El Cavet) from southern France and the Mediterranean coast of Iberia are compared with eight sites (3 caves and 5 open air sites)(Mala Triglavca, Grotta San Michele, Nakovana Cave, Danilo, Pokrovnik, Colle Santo Stefano, Fondo Azzollini and Ciccotto) from Italy and the Adriatic seashores with the same selection criteria.

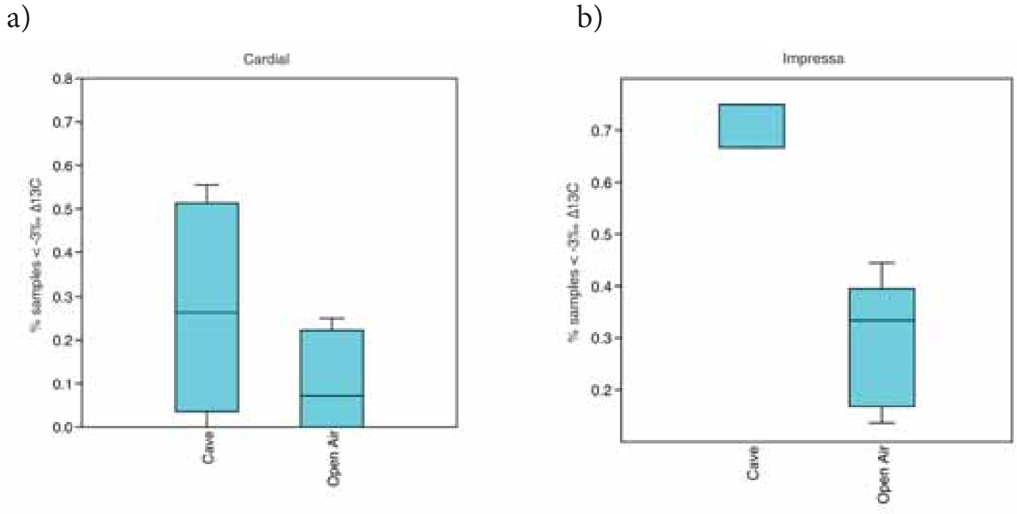


Figure 106: Box plots comparing the incidence of dairy products in caves and open-air sites from the Cardial (a) and Impressa (b) horizons.

Although, for the Cardial region, only caves are the sites with the highest amounts of recovered dairy products, both types of sites have been shown to also present from none up to 30% of pottery containing dairy products. A Student T test (N=9, T=1.2516, p=0.25731) indicates that the null hypothesis that Caves and Open Airs present the same means should not be rejected. Alternatively, the Italy and Adriatic region presents a significantly different picture where dairy residues in open air sites seem to be more frequent than their Iberian counterparts and caves contained the highest amounts of dairy isotopic signals (from 66% to 75% of the studied vessels). When trying to understand this pattern, the nature of the activities performed in the caves must be taken into account.

The occurrence of fireplaces suggest that certain caves might have been used as habitats (Bosch 2016b), foumier-type features indicate the possible existence of pens (Bergadà *et al.* 2003, Bergadà 1998a and b) and human bones suggest some cavities acted as sepulchres (Blasco *et al.* 2003, Oms *et al.* 2011, 2017).

The multiplicity of activities performed in this sites therefore suggest that, the possibility that pottery was acting as a grave good (Blasco *et al.* 2003), as a cooking tool or as a storage vessel, is translated into a set of organic residues which may or may not incorporate significant evidences for the production of dairy products. Therefore, the nature of the archaeological site do not seem to provide indications to help understand the dissociation between dairy products and non-ruminant adipose fats detected within this study.

8.2.3 Possible causes: composition of the herd

Alternatively, the range of animal products processed might be tightly linked with the range of species herded by the same group. The correlation between the number of ovicaprine remains in the studied archaeological sites with available data and the position of the residues in the porcine/dairy axis suggests that evidence for milk processing might have been linked with these particular species.

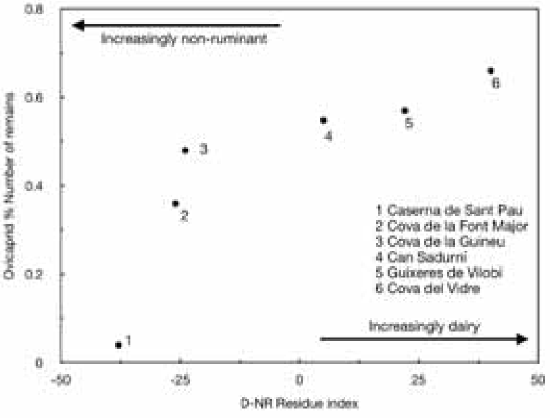


Figure 107: Correlation between the D-NR residue index and the percentage number of ovicaprid remains in Cardial sites. 1 Caserna de Sant Pau del Camp, 2 Cova de la Font Major, 3 Cova de la Guineu, 4 Guixeres de Vilobí, 5 Cova del Vidre, 6 Can Sadurní.

Identifying the species which could have been mainly used for milk production is not possible by solely studying the preserved fatty acids within the pottery matrices. Therefore, the fact that the expected amounts of dairy fats increased with higher quantities of ovicaprine remains must be treated with caution. Nevertheless, when studying the composition of the herds, the 7 sites with available data indicate that the amounts of ovicaprine and bovid remains are inversely correlated (Figure 107 and 108). In the light of this evidence, it would make less archaeological sense to expect dairy products to originate from the secondary exploitation of cattle even if their capacity of milk production per individual was significantly higher than smaller ruminants. Moreover, the study of the age at death (AtD) patterns in Can Sadurní (Saña *et al.* 2015) and Cova del Vidre (Saña *et al.* in press) are coherent with an ovicaprine exploitation adapted to the acquisition of milk, and the same strategy has been observed amongst sheeps and goats in the Postcardial layers within the Caserna de Sant Pau (Saña and Navarrete 2016) and in the impressa layers in Colle Santo Stefano (5840-5460, Italy)(Salque *et al.* 2012).

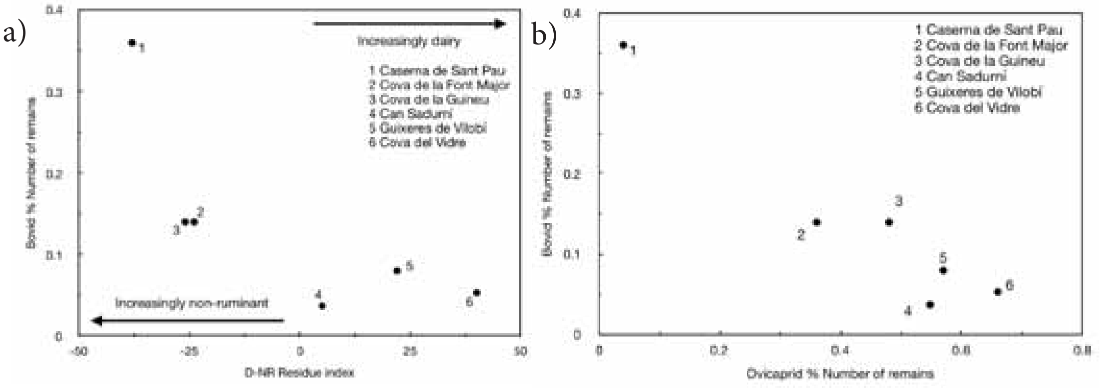


Figure 108: a) inverse correlation between the percentage number of bovid bone remains and the D-NR index in the Cardial period. b) inverse correlation between the percentage number of bovid and ovicaprid bone remains. 1 Caserna de Sant Pau del Camp, 2 Cova de la Font Major, 3 Cova de la Guineu, 4 Guixeres de Vilobí, 5 Cova del Vidre, 6 Can Sadurní.

Whether dairy practices might have been selectively practiced on Capra hircus or Ovis aries is a question impossible to explore given that only three of the studied sites reported different abundances for these two species (Table 54). The results for both the Cardial and Epicardial periods indicate that, in the studied sites, sheeps and goats were present in roughly the same proportions with the exception of Cova del Vidre (see Table 54). In this last case, the amount of organic residues evaluated through compound specific isotopic analyses is, by itself, too low (n=2). Therefore, further research incorporating sites with significant variation between the two species and the analysis of new pottery vessels will be needed to unlock the discussion of this issue with a minimal degree of certainty.

	5500-5000 cal BC.		**5000-4500 cal BC.**	
	Cova del Vidre	**Can Sadurní**	**Can Sadurní**	**Carrer Reina Amàlia**
Capra hircus	0.32	0.44	0.48	0.43
Ovis aries	0.68	0.56	0.52	0.57
Dairy intensity (D-NR)	40	6	4	4
Vessels analysed	2	4	5	10
Literature	Saña *et al.* in press	Saña *et al.* 2015 Spiteri *et al.* 2016		Navarrete and Saña 2015

Table 57: Relative amounts of sheep and goat bone remains as a percentage of the total quantity of ovicaprines.

The possible link between ovicaprids and dairying contrasts with Carrer Reina Amàlia (Epicardial), where the most abundant ruminant species are bovids (Navarrete and Saña 2015), and La Draga, not included in this study, where cattle AtD patterns are coherent with dairy production (Gillis *et al.* 2016). In this last site, the number of identified specimens and the number of remains suggest that ovicaprines would be present still in a higher percentage than cattle although when the potentially consumed biomass is taken into account, cattle would have been clearly the most prominent species (Saña 2011). Moreover, the possibility that milk from sheeps and goats could have been also obtained is not rejected by the researchers (Saña 2000). Although detailed reports on the organic residue analyses performed on pottery from La Draga have not been published yet, the researchers do indicate the presence of dairy products within the results (Gillis *et al.* 2016). Exemplified by La Draga and Carrer Reina Amàlia, this shows that, dairying practices linked with cattle herding might have been already present at the onset of the Neolithic in the western mediterranean in co-occurrence with strategies exploiting secondary products from goats or ovicaprines.

Patterns associating dairying and cattle herding have been mainly reported in Neolithic Britain, the Balkan mountain range and across the Middle East. Copley *et al.* (2005d), Ethier *et al.* (2017) and Evershed *et al.* (2008b) observed a significant increase of dairy fats linked with cattle herding which might be the result of the different composition of herds between the Mediterranean and Danubian neolithisation routes (Saña *et al.* 2013, Manning *et al.* 2013a, 2013b). Nonetheless, the data from this study suggests that the herding of seeps and goats could have been tightly linked with the production and consumption of dairy products in the western Mediterranean. This is coherent with other studied sites reporting dairy products in Italy and the Adriatic such as Colle Santo Stefano, where sheeps and goats were more than 50% of the amount of remains recovered (Salque *et al.* 2012, Radi and Wilkens 1989) and Mala Triglavca, where sheep and goat in the Vlaska group can be around 60% (Budja *et al.* 2013).

In conclusion, a general pattern linking dairy products and ovicaprines in the north-east of the Iberian Peninsula might have been complemented by other exploitation strategies where cattle would have been more prominent (ex: La Draga and Carrer Reina Amàlia). Nevertheless, data supporting this alternative approach is still scarce and suggests that the use of ovicaprids might have been the most practiced option. A more complete understanding of the prevalence or combination of both strategies is subject to the performance of more studies with higher sample quantities, but any future research in the Mediterranean will certainly need to assess a picture at least as complex as what has been suggested to date for the danubian neolithisation route (Manning *et al.* 2013a, 2013b).

In essence, the results obtained from this study provide further evidence supporting a need for analytical techniques which can distinguish between cattle and sheep/goat milk (Greenfield 2015). To confirm the hypotheses laid out here, the evaluation of protein constituents surviving as residues in pottery vessels (Hendy *et al.* 2018) seems one of the most promising venues.

8.3 Consumption practices regarding non-ruminant adipose fats.

To better understand the exclusive presence of non-ruminant carcass fats in sites where dairy is absent, trends and dynamics on non-ruminant product processing have been evaluated by comparing the expected median of non-ruminant products calculated by the Bayesian model and the amount of omnivore non-ruminant remains (mainly wild and domestic porcine remains) in the studied sites (Figure 109). The incorporation other wild species including *Meles meles*, *Felix silvestris* and *Vulpes Vulpes*, only detected in significantly minor amounts, does not affect the overall results and interpretation as major doubts could be expressed on its culinary value. Alternatively, *Oryctolagus cuniculus* remains were not taken into account as their reported compound specific isotopic values so far (Taché and Craig 2015, Choy *et al.* 2016) are significantly different than the archaeological data.

The situation in this case also presents a certain degree of complexity. Initially, there is no apparent correlation between the amount of non ruminant remains and their lipid counterparts. Nevertheless, several factors have to be taken into account. When observing Cova del Vidre (6), Can Sadurní (5), Cova de la Guineu (3) and Caserna de Sant Pau (1), a general trend can be defended. It could be argued that, within these sites, increasing quantities of non-ruminant animal bones were accompanied by increasing quantities of lipids coherent with non-ruminant adipose inputs in ceramic vessels. In the case of Cova de la Font Major and Cova del Vidre its values could be subjected to significant variation as only two samples from each site could be measured. Therefore, their values must be treated with caution. Alternatively, Guixeres de Vilobí (7) and Carrer Reina Amàlia (4) were calculated after averaging a higher amount of samples but

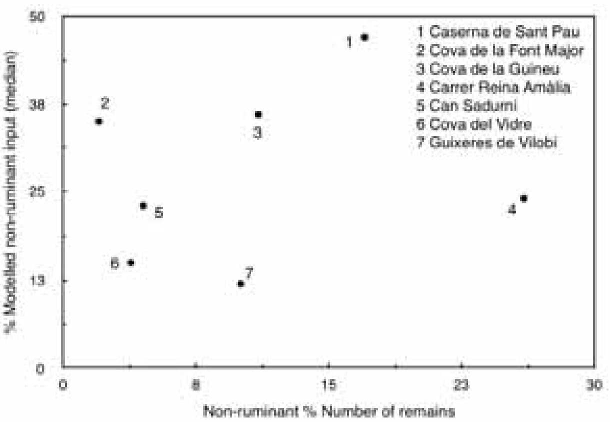


Figure 109: Percentage input from non-ruminant fats (median) as the result of bayesian modelling and the pecentage number of non-ruminant bone remains in the analysed archaeological sites.

present significantly lower quantities of non-ruminant adipose lipids although the amounts of omnivore non-ruminant remains in the sites suggests their presence in pottery cooking practices should be higher. Nonetheless, these two sites are characterised by the existence of domestic structures and evidences supporting permanent settlements, which is not the case of Can Sadurní, Cova de la Guineu and Caserna de Sant Pau. Had caves been used as places for herd stalling, there would be a significant functional difference between the sites not following and following a direct correlation between lipid residues and animal bones.

Neverhteless, more data on faunal remains and lipid residues are needed to fully understand porcine consumption practices across different types of sites as this is still not a clear trend. In the light of the available data, the possibility that open air sites with a strong domestic component (Guixeres de Vilobí and Carrer Reina Amàlia) could have repeatedly engaged in pork cooking techniques which did not involve the use of pottery (such as spit roasting) should not be rejected. To further explore the possible cooking techniques used by the first peasants and shepherds, molecular markers indicating food surpassed certain heat thresholds provides relevant information.

8.4 High temperature markers.

Where in-depth mass spectrometric studies were carried out (Can Sadurní, Cova del Vidre, Caserna de Sant Pau, Cova de Sant Llorenç and Carrer Reina Amàlia) multiple heat indicators were detected in several potsherds. This includes one sample (CV107) from Cova del Vidre, 4 samples (5, 9, 10 and 12) from Can Sadurní and 8 samples (9734, 9674, 9470, 9092, 8995, 8726, 6695 and 6405) from Carrer Reina Amàlia. The presence of ω–(o-alkylphenyl) octadecanoic acids would be indicative of the heating of polyunsaturated fatty acids of 18 carbons in its chain such as linoleic acid (C18:2) and linolenic acid (C18:3). Detection of cyclopentyl saturated octadecanoic acids would be coherent of the heating of monounsaturated fats (oleic acid, C18:1) and, finally, detection of the K31, K33 and K35 long chained ketones has been shown to correlate with the protracted heating of saturated stearic and palmitic acids. The range of recovered molecules therefore accounts for the majority of the fatty acids which can be found in animal fat triacylglicerides but their formation would not be expected under temperatures below 250ºC approximately. This threshold is not compatible with cooking practices such as boiling, stewing or any type of strategy which involves the use of water or water rich products as the energy needed to boil H_2O will keep the cooking temperature just under 100ºC. The detection of molecules originating at, at least, twice this temperature suggests other events would have affected the residues.

Before an archaeological explanation can be explored, the possibility that accidental or provoked fires might have altered the nature of the residues after pottery had been discarded must be taken into account.

In the Case of Carrer Reina Amàlia, although a certain amount of samples were found around a hearth, there were no evidences of post-depositional fires affecting layers 95 and 59. Markers for heating could only originate from the effects of the nearby hearth in layer 95 assuming that pottery sheds were already deposited and discarded around the fireplace when that one was still active. This possibility seems rather unlikely as heating markers are equally spread between samples found close and further away from the combustion structure.

In the case of Cova del Vidre, sample 107 was located in square K13, which is not close to any of the Neolithic hearths detected in the cave nor any other layers presenting evidences of combustion (Bergadà 1995). In consequence, there is no archaeological evidence in this case supporting the existence of post-depositional events which could have created the detected heating markers.

In the case of Can Sadurní, samples 9 and 12 were recovered from layer 18. This one (see section 5.2) contained significant evidence suggesting fire affected the artefacts in the layer shortly after its deposition. Micromorphology and heat fractures in lithic tools have been used to suggest temperatures would have been below 300/350ºC, which would be higher than the lower threshold observed for ω–(o-alkylphenyl) octadecanoic acids and cyclopentyl octadecanoic acids. Complementarily, the study performed by Spiteri (2012) highlighted the presence of long chained ketones in sample 13, which was also found in layer 18. Alternatively, samples 10 and 5, recovered from layers 13 and 10b respectively, were both deprived of any significant markers of postdepositional fires.

In conclusion, although the postdepositional formation of heating markers can not be ruled out for samples 9 and 12 in Can Sadurní, the remaining vessels recovered from the same site, from Cova del Vidre and from Carrer Reina Amàlia would have possibly seen the formation of said molecules due to other factors.

Intentional placing of the contents of pottery vessels at temperatures higher than 250ºC might occur due to a range of uses not necessarily motivated by the preparation of foodstuffs. The reuse of cooking vessels as metallurgical tools could be contemplated but is not coherent for the Early Neolithic period. Furthermore, when using pottery for illumination, all analyses of prehistoric lamp fuels so far do not report the presence of said pyrolithic products (de Beaune 1987, Copley *et al.* 2005e, Evershed *et al.* 1997b, Kimpe *et al.* 2001, Garnier *et al.* 2009, Mottram *et al.* 1999, Solazzo and Erhardt 2007) except for the ω-(o-alkylphenyl) alkanoic acids detected in late Mesolithic "blubber" lamps, which were only interpreted as markers for marine products (Heron *et al.* 2013). Finally, an additional possibility would be the intentional firing of the vessels as a cleaning mechanism by burning the embedded organic matter. Nevertheless, was this a generalised practice, it would be more commonly found across all pottery vessels, nevertheless it has been never reported regarding organic residue analysis. Its results could be assumed to be similar to firing pottery again, which, instead of creating pyrolithic products, results in the carbonisation of the available organic matter previously mixed with the clay (Drieu *et al.* 2019).

In consequence, the possibility that dairy (sample 107 from Cova del Vidre), and adipose fats (samples 9 and 12 from Can Sadurní and samples 9092 and 8726 from Carrer Reina Amàlia) were submitted to elevated temperatures as part of the cooking process seems to be one of the most plausible explanations. Experiments validating the formation of long chained ketones (Raven *et al.* 1997, Evershed *et al.* 1995a), ω-(o-alkylphenyl) alkanoic acids (Hansel *et al.* 20047, Evershed *et al.* 2008a) and cyclopenyl fatty acids (Dobson *et al.* 1996a and b) have been performed in conditions analogous to cooking. The conscious submission of foodstuffs to said ranges of temperatures could be coherent with frying, baking or, in some cases, accidents resulting in overcooking food. Moreover, these hypotheses are coherent with the archaeological record in Carrer Reina Amàlia (González *et al.* 2011). The presence of a combustion structure tentatively identified as an oven dedicated to pottery firing does not preclude the possibility that it could have been also used for the transformation of foodstuffs at temperatures higher than 100ºC.

Although evidence for the cooking of animal terrestrial products seems to be widespread amongst the analysed residues, other evidences supporting the presence of plant residues and the possible incidence of marine products must also be discussed.

8.5 Plant products

Although triacylglycerols from plants are usually dominated by unsaturated components (ex: C18:1, C18:2), these are usually absent or greatly reduced from archaeological residues due to the occurrence of significant oxidation though time (Dunne *et al.* 2016). The characterisation of lipids from cereal seeds is severely limited by the scarcity of chemically stable lipids (Colonese *et al.* 2017a), which are at least tenfold lower than animal-based food items (Hammann and Cramp 2018). Furthermore the incorporation of plant lipids such as alkylresorcinols in clay matrices has been shown to increase significantly when plant products are mixed with animal fats (Spiteri 2012, Hammann and Cramp 2018), a phenomenon which promotes the detection of mixtures of both types of products, but also facilitates the masking of plant biomarkers by the greater amount of animal fats usually absorbed. Therefore, the detection of plant products within archaeological organic residues in pottery has been usually focused on the identification of rarely occurring fatty acids (ex: ricinoleic acid) and other types of compounds such as phytosterols, esters and alkanes from waxes and resin di- and triterpenoids (ex: Heron *et al.* 2016).

When discussing resins, the study of prehistoric adhesives has been hitherto based on the detection of birch bark tar in European sites such as Giribaldi, Podrî l'Cortri (Regert *et al.* 2004), Makriyalos or Paliambela (Mitkidou *et al.* 2008) or possible bitumen which would have arrived at the region thanks to long distance networks (Chalain: Regert *et al.* 2004). Nevertheless, as it has been previously discussed, the incidence of birch (*Betula* sp.) in the Iberian Peninsula is significantly limited as this species is usually found in colder environments. Pollen records from cores in Vall d'en Bas, Banyoles and Torroella de Montgrí suggests only minimal amounts of this species could have been present at the onset of the Neolithic. Furthermore, minor amounts of *Betula* sp. charcoal remains have only been found in a handful of Early Neolithic sites including Cova 120, Codella and Balma del Serrat del Pont, which are all placed in the mountainous region of La Garrotxa (Revelles *et al.* 2018). When looking at other pollinic cores and charcoal evidences closer to the region where the studied sites are placed, birch is completely absent. There are no traces of this tree in the pollen records from Barcelona (Riera *et al.* 2016), or in anthracologic studies from any of the sites included in this research. (ex: Mensua and Piqué 2008).

Alternatively, other resin producing plants include conifers such as *Pinus* sp. or *Juniperus* sp. Both types are usually significantly present in the early neolithic pollen records (Revelles *et al.* 2018, Riera *et al.* 2016), Juniperus sp. charcoals were found in El Cavet, Cova de la Font Major or Can Sadurní and carbonised remains from *Pinus* sp. (ex: *halepensis*) have also been detected in Barranc d'en Fabra, El Cavet, Cova de la Font Major and Can Sadurní. As previously discussed, their resins are easily distinguishable given the singular presence of labdane-type diterpenoids in *Juniperus* sp. exudates.

Pistacia lentiscus is another source of resin, commonly known as mastic, which was widely used in antiquity in the eastern Mediterranean (Stern *et al.* 2003). It is composed of a distinguishable range of characteristic triterpenoids which makes its differentiation from the other resin producing species easy. Its seeds have been detected in sites such as Cova de Sant Llorenç and Carrer Reina Amàlia (Antolín 2013), its charcoal has also been recovered from the Neolithic layers in Can Sadurní and, its pollen was identified in cores extracted from the Barcelona plain (Riera *et al.* 2016).

Therefore, except for birch bark tar, there is carpological, anthracologic and pollinic evidence supporting the availability of a significant range of resin producing plants in the region where the studied archaeological sites were placed. As previously reported, detection of mainly abietane diterpenoids in some vessels suggests that the role pottery had when managing this product was, at least until more data is produced on the subject, limited. In most cases, methyl dehydroabietic acid is detected alone or accompanied by 7-oxo dehydroabietic acid, both only in trace amounts. Given these results, it is difficult to ascertain whether said traces of resin would securely correspond with its anthropic use as firing contamination must be kept as a possibility (Reber *et al.* 2018). In another case, the quantity and range of abietane diterpenoids detected has been used to argue in favour of their anthropic origin. This is the case of an epicardial vessel (CS11) from Can Sadurní, where abietanes (dehydroabietic acid (methyl ester), 6-dehydrodehydroabietic acid, 7-oxodehydroabietic acid, 15-methoxy dehydroabietic acid and retene) accompanied by primaranes (isoprimaric acid methyl ester) suggests that the vessel could have contained a conifer resin. Although pine resin markers such as retene, 6-dehydrodehydroabietic acid, methyl dehydroabietic acid and 7-oxo dehydroabietic acid coud raise from firing contamination, other polyaromatic hydrocarbons detected in the experimental studies (Reber *et al.* 2018) were not present in the archaeological sample, namely 18/19-norabieta-8,11,13-triene and 1,2,3,4-tetrahydroretene. Moreover, 15-methoxy dehydroabietic acid and isoprimaric acid mehyl ester, which were not present in the firing experiments (Reber *et al.* 2018), were positively identified in this pottery sample. Therefore, firing contamination is not solely capable of explaining the range of conifer biomarkers detected. The presence of this type of resin amongst this epicardial vessel thus constitutes the oldest evidence hitherto for the use of resinous products combined with pottery in the north-east of the Iberian Peninsula. As discussed in the introduction, these results are coherent with a hafting adhesive from La Draga (Tresserras 2000b, Palomo *et al.* 2011) and results from the Early Neolithic layers in Cueva del Toro, in the south of the peninsula (Tarifa *et al.* 2019).

There are a range of equally possible uses which motivated the incorporation of resin into the pottery matrix of this epicardial vessel. Firstly, the pot could have been involved in the acquisition and transformation process of resin itself. It could have been placed below a tree's open would to collect oozing exudates or used to heat the collected product. Alternatively, the vessel could have stored resin and participated in the process of hafting lithic tools. Finally, resins in the vessel could be intentionally acting as a waterproofing agent or be the result of a repair. In any case, the co-occurrence of diterpenoids with fatty acids suggests that none of the aforementioned possibilities should be considered as the vessel's only purpose.

In conclusion, although the frequency of resins in the Mediterranean archaeological register is strikingly low, it seems that conifer resins might have been the preferred type in absence of birch bark tar just since the beginning of the Neolithic.

Regarding plant waxes, although there is supporting evidence for the presence of hydrolysed and well preserved esters in samples from Can Sadurní (CS14 and CS20), their location in layers 10b and 11a3 respectively date the vessels from the Middle Neolithic. Further evidence for plant products in this site includes the detection of ricinoleic acid in sample CS5, which, again, dates from the Middle Neolithic (layer 10b). Their detection proves the ability of the implemented techniques to distinguish these plant biomarkers, but they were completely absent in all the Early Neolithic samples. Phytosterols, odd over even alkane ranges and isotopic values coherent with C4 plants were equally absent in the studied Early Neolithic sties. Only 10 samples contained P/S ratios coherent with plant triacylglicerols (around 4 or higher), but extensive oxidation prevented the detection of further lipid compounds which could ascertain their nature as plant matter. These were samples 300/3 and CV56 from El Cavet, Samples SE09, SE12 and SEP3 from La Serreta, Sample CS12 from Can Sadurní, Sample FM84 from Cova de la Font Major, samples CGP4 and GCP5 from Cova de la Guineu and sample BF4 in Barranc d'en Fabra.

When reviewing the occurrence of docosenoic acid in nature, the USDA Food composition database does not show the presence of C22:1 (undifferenctiated) in any terrestrial animal products to be higher than 0.23%. Moreover, the products reporting the highest amount of this molecule are mustard oil and herring oil. Nevertheless, the interpretative value on the detection of docosenoic acid relies on the secure identification of its double bond position. While C22:1ω11 (cetoleic acid) is the major isomer in marine products, C22:1ω9 is the most common isomer found in plants (Heron *et al.* 2010) and substantially present in the seed oils of the Brassicaceae family (ex: radish, mustard and turnip) (Colombini *et al.* 2005). The detection of 13-docosenoic acid and 11-eicosenoic acid in archaeological residues has been usually confirmed by the identification of its erythro and threo dihydroxy fatty acids, which are indicative of the double bond position (Copley *et al.* 2005e). Tridecanedioic acid, an oxidation product indicative of the double bond position but not linkable to the original fatty acid chain length, was also detected in samples coherent with Brassicaceae seed oils (Copley *et al.* 2005e).

Docosenoic acid has been commonly detected in potsherds from Caserna de Sant Pau, but there is no chromatographic evidence supporting their presence in others sites such as Cova del Vidre. Furthermore, C22:1 is only intermittently detected in Can Sadurní or Cova de la Guineu but the fact that no dihydroxy fatty acids have been securely identified in any case challenges its value as plant a biomarker. Although its widespread presence in trace amounts across many samples suggests it could be related to post-depositional contamination issues, analysed soil from burial 19 and hearth 17 in Caserna de Sant Pau contained no detectable amounts of docosenoic acid. Moreover, tridecanedioic acid, which could have resulted from the oxidation of erucic acid (C22:1ω9), is commonly detected within the same samples presenting C22:1. Given that no significant biomarkers for marine products were detected, the possibility that C22:1ω11 was present seems highly unlikely. Therefore, the hypothesis that docosenoic acid originated from pottery use should not be fully rejected.

Although weeds from the Brassicaceae family could have easily inhabited the Neolithic Mediterranean climate (Schmidt and Bancroft 2011), evidence of its presence within the studied archaeological sites is unclear. As an example, out of more than 300000 seed and fruit remains found in La Draga only some samples was found to be similar to a possible Brassicaceae seed (cf. Brassicaceae and *Brassica cf. rapa*) (Antolín 2013:290, Antolín pers. comm.). Moreover, none of the anthracologic and carpologic information available from the studied sites indicates the presence of this family as oleaginous plants are very difficult to find in a carbonized state. Nevertheless, palinologic reports do include them in the general “weed” category (Revelles *et al.* 2017). Assessing the incorporation of plants from the Brassicaceae family into the everyday use of Early Neolithic farmers, therefore, remains an unclear issue which might be solved with further and detailed organic residue analyses.

In the light of this evidence, the detection of docosenoic acid presents significant interpretative challenges. Its widespread presence in Caserna de Sant Pau does not fit a context deprived of other plant biomarkers. Alternatively, its appearance in potsherds with P/S ratios higher than 4 (GIP5 in Cova de la Guineu and FM84 in Cova de la Font Major) co-occurs with the presence of C20:1 (gondoic acid) in GIP5 and C18:2 (linoleic acid) in FM84, which reinforces the hypothesis that the later two residues could have resulted from containing certain plant products. The presence of conflicting data, thus, demands further research into the possible origin of this molecule. For example, analytical approaches which can resolve its isomers such as DMDS derivatisations might be a way forward.

In conclusion, data on the use of pottery vessels to process or store plant products remains inconclusive under the light of organic residue analyses. Secure identification of a partial suite of its biomarkers could only be achieved in a small subset of the studied residues. Nonetheless, the abundance of plant macro and micro remains in the archaeological sites incorporated in this PhD and the usual success of other techniques more suitable for the recovery of plant evidences such as phytolith analyses indicates that the importance of plant products in pottery vessels should not be rejected. Additional research focused on techniques lowering chromatographic and spectrometric detection limits such as selective ion monitoring (SIM) might be an adequate strategy to further evaluate the presence of plant biomarkers. This approach could also be combined with a more careful assessment of the degree of preservation of organic matter, which could be used to support the need of either acid based or dichloromethane based extractions.

8.6 On the absence of marine biomarkers

Certain molecules which have been considered marine biomarkers (phytanic acid and the C18 ω-(o-alkylphenyl) alkanoic acid) were detected in some of the analysed residues from this study. Although they can be found in fish oils, these two indicators are also present in ruminant adipose fats and other animal adipose tissues. Therefore, the detection of these two indicators alone can not be used to ascertain the presence of marine products within the studied samples. Furthermore, isotopic studies of the stearic and palmitic acids were never able to yield isotopic ratios which would plot within the expected values for marine products. Nevertheless, this data should not be utilised to reject the use of marine products by the first peasants and shepherds in this region. Ictiofaunal remains and net weights (Gómez *et al.* 2014b) recovered from sites including Caserna de Sant Pau, Guixeres de Vilobí, Carrer Reina Amàlia are complemented by a later assemblage of 161 fish remains and fishing tools from the Gavà mines (Bosch *et al.* 1999). This suggests that the exploitation of marine or freshwater fish was not absent in the Early Neolithic. Moreover, the abundance of marine seashells and the practice of cardial decorations certify that marine products were recognised and incorporated into the everyday life. This is also clear when looking at jewellery, which, amongst other materials, included marine seashells across the Neolithic.

Only within the analysed sites, evidence for seashell jewellery was proven by polishing from anthropic origin in the symphonial face of seahsells in El Cavet (Fontanals *et al.* 2008a), several seashell beads in Cova de Sant Llorenç (Borrell *et al.* 2016), a fragment of a bracelet made on *Glycymeris* sp. and two bracelet fragments from Can Sadurí. Later in the Epicardial period these evidences would be complemented by remains of raw materials used for the preparation of personal attire in Carrer Reina Amàlia (Nadal 2016) and 19 seashell necklace beads from Barranc d'en Fabra (Bosch *et al.* 1995a).

Isotopic dietary studies are of outmost importance when trying to understand the relevance of marine products in this period. Nevertheless, to date, although several projects are underway, no results have been published yet for the studied region. Available data includes only tentative $\delta^{13}C$ measurements performed alongside ^{14}C dates. None of the VIth and Vth millennia human individuals buried at Cova Bonica (Oms *et al.* 2017), Plaça Vila de Madrid (Pou *et al.* 2010) and Carrer Reina Amàlia presented $\delta^{13}C$ isotopic values higher than -17‰, which might support the idea that correction of ^{14}C dates due to marine reservoir effects might not be necessary. In coastal archaeological sites south of the Ebro River such as Cova dels Diablets and Costamar (Salazar-García 2009, Salazar-García 2014a) palaeodietary studies showed no evidence for the consumption of freshwater or marine residues in significant quantities. In consequence, and taking all available data into account, the existence of evidence supporting a significant consumption of marine products within vessel residues is lacking in this study and in other cases across the Mediterranean (Spiteri 2012, Tarifa *et al.* 2019). However, its absence in pottery does not reject the possibility that fish was exclusively roasted on sticks next to the fireplace but, to date, the only compelling evidence for the dietary consumption of marine products is constrained to cockles, limpets and other simmilar species (Bosch *et al.* 1995a, Nadal *et al.* 2015) whose weight in the overall diet remains an open question. In conclusion, although the relevance of the sea within the symbolic sphere in the Early Neolithic is undeniable, further palaeodietary studies, more detailed analysis of the diastereoisomers of phytanic acid in pottery residues, and a greater care to detect evidence for the consumption of marine products by fieldwork archaeologists will be needed to shed new light onto this issue in the future.

8.7 Fungal activity and fermentation

Beyond heat, fermentation is one of the cooking techniques which could have been widely in place at the onset of the Neolithic. There is a significant body of work evaluating the possible significance of the introduction of fermented beverages amongst prehistoric societies (ex: Hayden *et al.* 2012, Guerra 2008) both linked to the beginning of the Neolithic and to later in the Bronze Age. Moreover, fermentation processes significantly reducing the amount of lactose in dairy products could be expected in periods were the gene for lactase persistence was completely absent (Olalde *et al.* 2018, Lacan 2011, Sverrisdóttir *et al.* 2014). Nevertheless, the study of molecular markers used to identify fermentation (calcium oxalates, Ergosterol, bacterial hopanoids (Correa-Ascensio *et al.* 2014)), remain a challenging field (Salque *et al.* 2016) as post-depositional fungi and bacteria could yield similar results. As previously indicated, Ergosterol has been detected in several experimental studies after vessel burial (Hammann and Cramp 2018, Spiteri 2012, Dudd *et al.* 1998). Nevertheless the fact that this molecule (Isaksson *et al.* 2010, Namdar *et al.* 2015) and its degradation products (Shoda *et al.* 2017, Colonese *et al.* 2017a, Reber *et al.* 2015) have been detected in several archaeological cases indicates that further discussion on the possible anthropic origin of the compound is needed.

Although the GC and GCMS analyses performed in this study would be able to detect Ergosterol and bacterial hopanoids, these molecules were absent in all archaeological cases. Nonetheless, Ergosterol and several ergosteroids could be found in one experimental sample. After the development of a modern beer recipe inspired in the findings at Cova de Can Sadurní, the preparation of this beverage was performed on a replica vessel. Malt, water, Thyme, Artemisia, strawberry and honey were mixed and left to ferment inside the Cave for some time[2]. The residue left after the fermentation process consisted of visible non-carbonised food crusts which started to slowly degrade. The sampling of this pot implicated the removal of tiny amounts of visible residues and drilling roughly 2g of pottery from the base and the rim of the vessel.

---

[2] The full recipie and the experimental process can be viewed in the national geographic webpage

Organic matter was extracted following a solvent extraction with DCM:MeOH 2:1 v:v, derivatised with BSTFA and analysed under High Temperature Gas Chromatography-Mass Spectrometry. The resulting chromatogram (figure 110) was a complex mixture of a wide range of organic molecules. Given that the vessel had not been subjected to extensive oxidation and hydrolytic processes, the analytical result contained evidences for almost all of the ingredients mixed in the vessel. As presented in the slide, the plant material used in the preparation of beer (malt, aromatic herbs and strawberries) could be linked with a fatty acid profile containing plant specific molecules such as ricinoleic acid and β-sitosterol and a high P/S ratio (2.7). It is important to notice that, despite the minimal amount of time it had passed since the vessel had been used compared with thousands of years of burial in archaeological cases; lipids had already started to degrade (Figure 110).

Mono-, di- and polisaccharides tentatively detected across the chromatogram could originate from a combination of the sugars in honey and strawberries and the sugars released by the malting process. Given that the analysis was performed with parameters scanning for a wide range of possible molecules, there was no analytical resolution available to distinguish them further.

The detection of ergosterol and ergosta-4,6,8(14),22-tetraen-3-one (a fungal metabolite (Seitz and Paukstelis 1977), could be interpreted as a marker for the presence of the agents needed for fermentation. Although the compound could also be generated by other fungi and microorganisms living in soil, organic matter inside the clay matrix would have been usually more protected from external contamination than visible external residues. Therefore, although

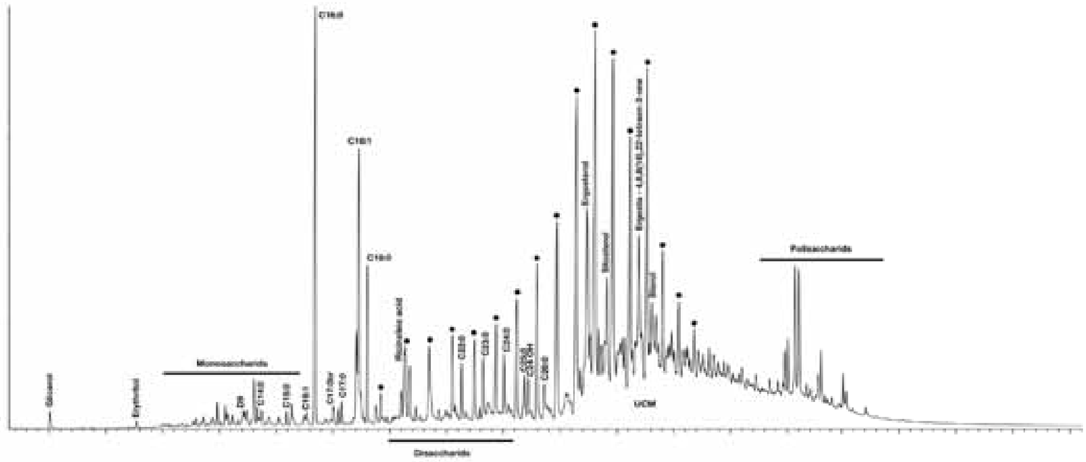


Figure 110: Chromatogram of the analysed experimental sample. · Alkanes.

the contamination hypothesis must not be completely ruled out, the presence of ergosterol could also be explained by the fermentation process needed to produce beer.

Moreover, the significant amount of linear alkanes detected presents a complex interpretation. A petrogenic origin seems unlikely due to the complementary absence of phytane, pristane, hopanes and stearanes but the absence of preference between odd and even chains is not coherent with plant waxes either. In light of the presence of Ergosterol, the possibility that defunctionalisation of previous polar linear compounds via microbiological activity should not be fully ruled out.

In conclusion, the analysis of the organic remains found in an experimental pot used to brew beer contained a range of products that would have certainly degraded over time and would thus not be found in archaeological pottery. These would be mainly carbohydrates. Nonetheless, it also indicated that this activity would certainly produce residues with recalcitrant chemical compounds which have already been found in archaeological pottery: free fatty acids, ricinoleic acid, ergosterol, other sterols and a wide range of alkanes. Although none of these elements is unique and specific for beer, their combined detection in archaeological pottery would imply that the presence of fermentation products should not be ruled out.

9 Conclusion

A thorough discussion of the results obtained from the analysis of 190 samples from the Cardial and Epicardial Neolithic across the north-eastern Iberian seashore has been able to detect significant correlations and cast new light into the economic strategies practiced by the first farmers and herders in the western Mediterranean.

Recovered data from organic residue analyses performed on 190 Cardial and Epicardial pottery sherds yielded significant lipids in 156 cases (82%) which illustrates the presence of terrestrial animal foodstuffs within pottery. However further research will be needed to positively identify plant and marine remains. Arguably, the absence of proof does not constitute proof of absence as biomarkers for plant and marine products would be more susceptible to degradation. Even if plant and marie foodstuffs were effectively cooked in pottery, their signal might not have survived in the manner investigated through organic residue analysis.

Where possible, differences in use between pottery types across both the Cardial and Epicardial periods were explored. Although some differences in the Cardial period between appliqués and impressed decorations could be in place in La Serreta, these same categories did not show any significant variation in contemporary layers in Cova de la Guineu. In sites from both periods (La Serreta and Carrer Reina Amàlia), the firing conditions seemed to affect the total lipid extract as vessels cooked in oxidising atmospheres presented lower amounts of residues. In Barranc d'en Fabra, where diameter estimates were available, higher quantities of lipids might have been present in wider Epicardial vessels. Variation between vessel size and the characteristics of the residues could also be tentatively identified in Carrer Reina Amàlia, where thinner vessels (lower than 10mm) were the only ones containing dairy products or isotopic signals closer to the dairy threshold. Alternatively, biomarkers for temperatures higher than around 250°C tended to be in thicker pots. Although specific correlations between pottery and residue characteristics have been possible in both the Cardial and Epicardial periods, its relevance should be treated with caution. Rather than being a widespread tendency, these seem to be constrained at the site scale and, in this situation, future analyses might provide data which could significantly alter these interpretations.

When the type of site has been taken into account, sites with the strongest evidences of human open-air settlement (Guixeres de Vilobí and Carrer Reina Amàlia) presented abnormally low quantities of non-ruminant fats in the vessels. This suggests the possible existence of other cooking techniques which would not use pottery, such as spit-roasting. Furthermore, caves and open air sites, although found to differ in the quantity of dairy residues detected in the central Mediterranean, did not seem to significantly diverge in the northeast of the Iberian Peninsula.

A comparison between the three studied regions (Barcelona plain, Garraf and Penedes and South and Ebro valley) shows a slight tendency towards mixtures incorporating more non-ruminant adipose fats in the central coastline (Barcelona plain) rather than the south-west (Ebro valley). This tendency has only been detected in the Cardial period as it could not be confirmed in Epicardial samples. Overall, organic residues in pottery present a significant continuity between the second half of the VIth millennium and the first half of the Vth millennium cal BC.

The increasing amount of ovicaprine remains in sites yielding dairy fats supports the existence of three main "culinary practices" explained by the (1: Cova del Vidre, Can Sadurní, El Cavet, Guixeres de Vilobí) presence of dairy products and ruminant carcass fats accompanied, when data is available, by significant quantities of ovicaprine remains and, in one case, (Carrer Reina Amàlia), cattle remains; (2: Caserna de Sant Pau, Cova de la Font Major, La Serreta and Cova de la Guineu) the presence of non-ruminant and ruminant carcass fats in sites with varied amounts of non-ruminant bone remains and (3: El Molló, Barranc d'en Fabra) the existence of two sites where, neither dairy nor non-ruminant carcass fats were detected. Finally, additional isotopic data from Cova de Sant Llorenç and Cau de les Guilles will be necessary in order to obtain indications regarding to which group they might have belonged.

Beyond terrestrial animal products, the presence of diterpenoids coherent with conifer resins is the clearest evidence for the incorporation of plant organic matter into clay matrices. Although in most cases their nature as an archaeological residue is unsure, the presence of abietane and primarane-types in at least one vessel from Can Sadurní provides a secure evidence for the exploitation of conifer resins in the Epicardial Neolihtic in comparison with other available species such as *Juniperus* sp. or *Pistacia lentiscus*. Regarding marine products, a thorough analysis of the molecular and isotopic biomarkers available for lipid organic residue analysis did not yield any evidence supporting their use involving pottery vessels. Nevertheless, the punctual detection of fish remains in the coast, the widespread presence of marine seashells in archaeological sites, and their use as tools to decorate pottery and manufacture jewellery supports a scenario where marine products could have been still a minor component of the diet. To date, paleodietary studies from human bones around the studied area do not contradict this interpretation.

Regarding cooking strategies, the detection of significant amounts of fats close to the rim of the studied vessels suggests boiling and cooking with major amounts of water could have been widespread in this moment. Detection of biomarkers from heating events surpassing 250°C points at the possible existence of other cooking strategies such as frying or baking. Nevertheless, despite of its positive evidence in modern experiments, no compelling evidence for fermentation could be identified in archaeological vessels. In conclusion, the range of molecular and isotopic evidences gathered so far point at the existence of the range of cooking methods which are nowadays found in most traditional cuisines. Moreover, the association between ovicaprine remains and dairy residues supports the hypothesis that the Mediterranean neolithisation pathway would have developed specific traits as a result of their interaction with the landscape. In the case of the procurement of milk, additional species such as sheep or goats might have been managed for their secondary products as well. Therefore, this suggests the existence of Mediterranean-specific culinary practices and the presence of a mature cooking toolset already around at least 7500 years ago.

The results and interpretations arising from this PhD open and revisit certain limitations and future challenges of organic residue analyses in pottery. In most cases, the patterns detected by this study could benefit from larger sample sizes per site. This might be a difficult challenge in small sites for two main reasons. In some cases the minimum number of vessels calculated was effectively sampled, this was the case in La Serreta and Cau de les Guilles. In other cases the degree of fragmentation and preservation of the remains was considerably low. The remaining potsherds would just not be suited for organic residue analysis given their low weight and the lack of relevant ceramic information; this seems to be the case for Barranc d'en Fabra and Cova de Sant Llorenç. The advancement of new and more finely tuned techniques may allow further sampling in the future. Certain studied sites contained a number of analysis more coherent with a pilot research than a full study and further sampling would surely significantly impact the interpretations performed for them, this could be the case of Cova de la Font Major, Cova del Vidre and Berenguer de Palou. Further sampling in sites currently under excavation might be also advisable as more pottery becomes available, this would affect Guixeres de Vilobí, Cova de la Guineu, Can Sadurní or El Cavet.

In the cases where the analyses resulted in rich and complex chromatograms, its interpretation might be significantly enriched by sampling the same fragment further and performing solvent extractions to confirm the presence of complex structures vulnerable to hydrolysis. This could be performed in cases suggesting the presence of wax esters from Can Sadurní. Furthermore, samples identified as ruminant dairy fats which present a total lipid extract suggesting triacylglycerols (TAG) might have survived intact could be analysed under a solvent extraction to attempt at differentiating between bovid and ovicaprid TAG profiles.

Furthermore, although the correlation between bones and lipid residues is highly informative, it does not constitute a full proof of the species of animals which could have been used for dairying. A path to overcome this limitation would be the development of new analytical strategies capable of distinguish dairy residues at the species level. However, the limited amount of information preserved in lipids may prevent this differentiation. The use of other more informative molecules, for example proteins or ancient DNA, might constitute a solution; nevertheless these are water soluble and therefore significantly more prone to degradation. Although recent advances seem promising, significant work will be needed to select and analyse samples amenable for this kind of analysis and control for possible modern contamination.

When evaluating the knowledge about the earliest Neolithic societies gained in this research, it seems clear that the tendencies and patterns tentatively detected in the modern catalan coast could be tested further in other Iberian territories. This peninsula incorporates a wide range of geographic domains which could have resulted in different culinary adaptations. Studies on the full span of the Ebro valley, the Pyrenees or the Baetic mountain range, the Cadiz gulf or the Gibraltar straight might help better paint the complex picture that is the neolithisation of the Iberian Peninsula.

Beyond the geographic domain, organic residue analysis might be capable to inform about the profound socioeconomic changes that underwent the Early Neolithic societies during the second half of the Vth millennia. The increment on the diversity of archaeological contexts such as necropolis and megalithic tumbs and significant changes in decorative styles might be accompanied by shifts in other aspects of society such as the culinary practices. In these periods, the postcardial and middle Neolithic, the possibility to compare residues from grave goods and settlements and the availability of extensive paleodietary studies presents a unique opportunity for lipids to inform not only culinary but also ritual practices and to assess whether and how they could have changed by comparing them to the Early Neolithic.

At the end of this PhD it is also clear that more work will be needed regarding the identification of plant and marine products. Additional analytical steps including operating mass spectrometers in selective ion monitoring (SIM) could be used to target specific biomarkers and specific chromatographic programs could be used to evaluate phytanic acid diastereoisomers. Furthermore, the detection of non-edible organic materials such as resins will certainly help better understand the first uses of pottery beyond cooking in the first Neolithic. So far, detection of diterpenoids clearly attributable to ancient residues might be challenged by possible contamination from hearth smoke. Hopefully, the development of specific analytic tools and guidelines for its secure detection will certainly help unravel the role of one of the most necessary materials for everyday prehistoric life and yet one of the most elusive substances in the archaeological record, glues.

Finally, it can be argued that this PhD constitutes a significant contribution to the available knowledge regarding the first culinary practices involving pottery in the north-east of the Iberian Peninsula. Data revealed in this research has successfully detected the presence of ruminant dairy residues linked to ovicaprine herding and has assessed the variation of the lipid residues across pottery types, site types, space and time. As a consequence from this analysis it has been suggested that the previous proposed link between dairying and caves in the central Mediterranean might not be in place in the west. In essence, a gap in the knowledge about the first uses of pottery in the last territories of the Mediterranean neolithisation route has been filled and new future lines of research have been identified.

10 Bibliography

A

Ache, M. (2011). Els artefactes macrolítics dels nivells neolítics de la cova de Can Sadurní. In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació.* (pp. 129–140). Edar. Arqueología y Patrimonio.

Ackman, R. G., & Hooper, S. N. (1968). Examination of isoprenoid fatty acids as distinguishing characteristics of specific marine oils with particular reference to whale oils. *Comparative Biochemistry and Physiology*, 24(2), 549–565. https://doi.org/http://dx.doi.org/10.1016/0010-406X(68)91008-6

Aillaud, S. (2002). *Field and Laboratory Studies of Diagenetic Reactions Affecting Lipid Residues Absorbed in Unglazed Archaeological Pottery Vessels.* PhD thesis. Shcool of Chemistry. University of Bristol, Bristol.

Albarella, U., & Serjeantson, D. (2002). A passion for pork: meat consumption at the British Late Neolithic site of Durrington Walls. *Consuming Passions and Patterns of Consumption*, 33, 49.

Alcalde, G., Bosch, À., & Buxó, R. (1991). L'assentament neolític a l'aire lliure de Plansallosa (la Garrotxa). *Cypsela*, 1991, Núm. 9, p. 49-63.

Alcalde, G., Molist, M., & Saña, M. (1999). Avance preliminar sobre la dinámica de las ocupaciones neolíticas documentadas en el yacimiento de la Bauma del Serrat del Pont (La Garrotxa, Catalunya). *SAGVNTVM Extra*, 2, 337–342.

Alcalde, G., Molist, M., Saña, M., & Borrell, F. (2002). *Procés d'ocupació de la Bauma del Serrat del Pont, La Garrotxa, entre 5480 i 2900 cal AC*. Museu comarcal de la Garrotxa.

Alcalde, G., Barberà, L. C., de Haro, S., Lladó, E., Saña, M., & Tornero, C. (2008). Dinámica de asentamiento en la zona volcánica de la Garrotxa (Catalunya) durante es neolítico antiguo. In M. S. Hernández, J. A. Soler, & J. A. López (Eds.), *IV Congreso del Neolítico Peninsular* (Vol. 1, pp. 216–220). Alicante: Museo Arqueológico de Alicante.

Allué, E. (2003a). Aspectos ambientales y económicos durrante el Neolítico Antiguo a partir de la secuencia antracológica de la Cova de la Guineu (Font Rubí, Barcelona). In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico en la Peninsula Ibérica* (pp. 53–61). Santander: Monografías del Instituto Internacional de Investigaciones Prehistoricas de Cantabria.

Allué, E. (2003b). Les anàlisis antracològiques a Tarragona i el seu entorn: l'interès per l'estudi de la vegetació del passat i la utilització dels recursos forestals. *Butlletí Arqueològic*. Reial Societat Arqueològica Tarraconense, 25, 5–17.

Alonso, F., Cabrera, V., Chapa, T., & Fernandez-Miranda, M. (1978). Índice de fechas arqueológicas de C-14 para España y Portugal. In M. Almagro (Ed.), *C14 y Prehistoria de la Península Ibérica* (Vol. 77). Madrid: Fundación Juan March.

Anfruns, J., Majó, T., & Oms, J. I. (1995). Estudio preliminar de los restos humanos neolíticos pàrocedentes del yacimiento de la calle Sant Pau (Barcelona). In *Congreso español en Antropologia Biológica* (pp. 43–51).

Antolín, F. (2013). *Of cerecals, poppy, acorns and hazelnuts. Plant economy among early farmers (5500-2300 cal BC) in the NE of the Iberian Peninsula. An archaeobotanical approach*. PhD Thesis. Departament de Prehistòria. Universitat Autònoma de Barcelona, Barcelona.

Antolín, F., Ache, M., Bergadà, M. M., Blasco, A., Buxó, R., Edo, M., ... Villalba, M. J. (2011). Aproximació interdisciplinària a l'acció del foc en les inhumacions i aixovars del Neolític antic cardial de Can Sadurní (Begues, Baix Llobregat). In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació*. (pp. 151–158). Edar. Arqueología y Patrimonio.

Antolín, F., Buxó, R., Jacomet, S., Navarrete, V., & Saña, M. (2014). An integrated perspective on farming in the early Neolithic lakeshore site of La Draga (Banyoles, Spain). *Environmental Archaeology*, 19(3), 241–255.

Antolín, F., Navarrete, V., Saña, M., & Gassiot, E. (2018). Herders in the mountains and farmers in the plains? A comparative evaluation of the archaeobiological record from Neolithic sites in the eastern Iberian Pyrenees and the southern lower lands. *Quaternary International*, 484, 75–93. https://doi.org/10.1016/j.quaint.2017.05.056

Aura, J. ., Jordá, J., Pérez, M., & Rodrigo, Mo, J. (2001). Sobre dunas, playas y calas. Los pescadores prehistóricos de la Cueva de Nerja (Málaga) y su expresión arqueológica en el tránsito pleistoceno-holoceno. *Archivo de Prehistoria Levantina*, XXIV.

B

Baeten, J., Romanus, K., Degryse, P., De Clercq, W., Poelman, H., Verbeke, K., ... Waelkens, M. (2010). Application of a multi-analytical toolset to a 16th century ointment: Identification as lead plaster mixed with beeswax. *Microchemical Journal*, 95(2), 227–234. https://doi.org/10.1016/j.microc.2009.12.005

Baeten, J., Deforce, K., Challe, S., De Vos, D., & Degryse, P. (2014). Holy Smoke in Medieval Funerary Rites: Chemical Fingerprints of Frankincense in Southern Belgian Incense Burners. *PLoS ONE*, 9(11), e113142. https://doi.org/10.1371/journal.pone.0113142

Baldellou, V. (1971). La Necropolis Prehistórica del "Barranc d'en Fabra" (Amposta). *Butlletí Arqueològic*. Reial Societat Arqueològica Tarraconense, 113–120, 41–49.

Baldellou, V. (1972). El yacimiento prehistórico del Roc d'en Sardinyà (Sant Genis de Vilassar, Barcelona). *Pyrenae*, 8, 41–52. Retrieved from http://www.raco.cat/index.php/Pyrenae/article/view/164865/242374

Baldellou, V. (1974). Los materiales arqueológicos de la "Cova Bonica" de Vallirana (Barcelona). *Ampurias: Revista de Arqueología, Prehistoria y Etnografía*, (36), 1–20.

Baldellou, V., & Mestres, J. (1981). Les Guixeres de Vilobí, hàbitat del Neolític Antic a l'aire lliure. In G. Camps, M. Llongueras, R. Marcet, M. A. Petit, & J. Rovira (Eds.), *El Neolític a Catalunya. Taula rodona de Montserrat*. (1a ed., pp. 69–74). Tortosa, Spain: Publicacions de l'Abadia de Montserrat.

Ballesteros, A. (2009). Restitució paleoambiental de l'entorn de la cova de la Font Major (l'Espluga de Francolí) mitjançant la palinologia i els sistemes d'informació geogràfica. *Aplec de Treballs*, (27), 71–80.

Barceló, J. A. (2008). La incertesa de les cronologies absolutes en arqueologia. Probabilitat i estadística. *Cypsela: Revista de Prehistòria i Protohistòria*, 17, 23–33.

Barnard, H., Ambrose, S. H., Beehr, D. E., Forster, M. D., Lanehart, R. E., Malainey, M. E., ... Yohe Ii, R. M. (2007). Mixed results of seven methods for organic residue analysis applied to one vessel with the residue of a known foodstuff. *Journal of Archaeological Science*, 34(1), 28–37. https://doi.org/http://dx.doi.org/10.1016/j.jas.2006.03.010

Barnard, H., Dooley, A. N., Areshian, G., Gasparyan, B., & Faull, K. F. (2011). Chemical evidence for wine production around 4000 BCE in the Late Chalcolithic Near Eastern highlands. *Journal of Archaeological Science*, 38(5), 977–984. https://doi.org/http://dx.doi.org/10.1016/j.jas.2010.11.012

Bartrolí, R., Bergadà, M., Cebrià, A., & Fontugne, M. (1992). El model microlaminar a la Catalunya meridional: aportacions del projecte de la Serra de Font-rubí (Alt Penedès). In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 34–37). Puigcerdà, Spain: Institut d'Estudis Ceretans.

Bastos, B. (2013). *Potential of lipid analysis on prehistoric Portuguese pottery.* MSc dissertation. University of Bradford.

Bayes, T. (1763). An essay towards solving a Problem in the Doctrine of Chances. By the late Rev. Mr. Bayes, F.R.S. communicated by Mr. Price, in a Letter to John Canton, A.M.F.R.S. *Phylosophial Transactions of the Royal Society*, 53, 370–418.

Bayless, T. M., Brown, E., & Paige, D. M. (2017). Lactase Non-persistence and Lactose Intolerance. *Current Gastroenterology Reports*, 19(5), 23. https://doi.org/10.1007/s11894-017-0558-9

Beja-Pereira, A., Luikart, G., England, P. R., Bradley, D. G., Jann, O. C., Bertorelle, G., ... Erhardt, G. (2003). Gene-culture coevolution between cattle milk protein genes and human lactase genes. *Nature Genetics*, 35(4), 311–313. https://doi.org/10.1038/ng1263

Bergadà, M. M. (1995). Estudio geoarqueológico de la secuencia Holocena de la Cova del Vidre (Roquetes, Baix Ebre, Tarragona). In M. Borrell, J. Bosch, & A. Ribes (Eds.), *I Congrés del Neolític a la Península Ibèrica*. (Vol. 1, pp. 65–72). Gavà/ Bellaterra: Rubricatum.

Bergadà, M. M. (1998a). Cueva de la Guineu. In *Estudio geoarqueológico de los asentamientos prehistóricos del Pleistoceno Superior y el Holoceno inicial en Catalunya* (Vol. 742, pp. 99–130). Oxford, UK.

Bergadà, M. M. (1998b). Cueva del Vidre. In *Estudio geoarqueológico de los asentamientos prehistóricos del Pleistoceno Superior y el holoceno inicial en Catalunya.* (Vol. 742, pp. 205–234). Oxford, United Kingdom.

Bergadà, M. M, Cebrià, A., & Mestres, J. (2003). Prácticas de estabulación durante el Neolítico Antiguo en Cataluña a través de la micromorfología: cueva de la Guineu (Font-Rubí, Alt Penedès, Barcelona). In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico en la Peninsula Ibérica* (pp. 187–196). Santander: Monografías del Instituto Internacional de Investigaciones Prehistoricas de Cantabria.

Bernabeu, J. (1989). La tradicion cultural de las ceramicas impresas en la zona oriental de la peninsula ibérica. *Serie de Trabajos Varios-Servicio de Investigacioń Prehistórica,* (86).

Bernabeu, J. (1999). Pots, symbols and territories: the archaeological context of neolithisation in Mediterranean Spain. *Documenta Praehistorica,* 26, 101–118.

Bernabeu, J., Orozco, T., Díez, A., Gómez, M., & Molina, F. (2003). Mas d'Is (Penàguila, Alicante): aldeas y recintos monumentales del Neolítico Inicial en el valle del Serpis. *Trabajos De Prehistoria*, 60(2), 39–59. https://doi.org/10.3989/tp.2003.v60.i2.80

Bernabeu, J., Molina, L., Esquembre-Bebiá, M. A., Oetega, J. R., & Boronat, J. de D. (2009). La cerámica impresa mediterránea en el origen del Neolítico de la península Ibérica. In *De Méditerranée et d'ailleurs...: mélanges offerts à Jean Guilaine* (pp. 83–96).

Bernabeu, J, García, P., Gómez, O., & Molina, L. (2011). El componente decorativo en las producciones cerámicas. *SAGVNTVM*, (Extra 12), 17–34.

Bernabó, L. (1956). *Gli scavi nella Caverna delle Arene Candide (Finale Ligure). Parte prima: gli strati con ceramiche. Campagne di scavo 1948-1950* (Vol. 2). Bordighera.

Berrocal, A. (2018). Las bases de nuestra cocina. In A. Palomo, R. Piqué, & X. Terradas (Eds.), *La revolución Neolítica. La Draga, el poblado de los progidios.* (pp. 89–92). Madrid, Spain: Museo Arqueológico Regional de la Comunidad de Madrid.

Bersaglieri, T., Sabeti, P. C., Patterson, N., Vanderploeg, T., Schaffner, S. F., Drake, J. A., ... Hirschhorn, J. N. (2004). Genetic signatures of strong recent positive selection at the lactase gene. *The American Journal of Human Genetics,* 74(6), 1111–1120.

Binder, D. (1991). *Une Économie de Chasse au néolithique ancien. La Grotte Lombard À Saint-Vallier-De-Thiey*. Monographie du CRA. Editions du CNRS.

Binder, D., Clop, X., Convertini, F., Manen, C., & Sénépart, I. (2010a). Les productions cerámiques du Néolithique ancien entre Provence et Catalogne. In C Manen, F. Convertini, D. Binder, & I. Sénépart (Eds.), *Premières sociétés paysannes de Méditerranée occidentale. Structures des productions céramiques.* (pp. 29–42). Toulouse: Société Prehistorique Française, Memoire LI.

Binder, D., Convertini, F., Manen, C., & Sénépart, I. (2010b). Les productions céramiques du Néolithique ancien: proposition d'un protocole d'analyse. In C. Manen, F. Convertini, D. Binder, & I. Sénépart (Eds.), *Premières sociétés paysannes de Méditerranée occidentale. Structures des productions céramiques.* (pp. 29–42). Toulouse: Société Prehistorique Française, Memoire LI.

Blasco, A, Edo, M., Villalba, M. J., Buxó, R., Juan-Tresseras, J., & Saña, M. (1999). Del cardial al postcardial en la cueva de Can Sadurní (Begues, Barcelona). Primeros datos sobre su secuencia estratigráfica, paleoeconómica y ambiental. In J. Bernabeu, T. Orozco. *II Congrés Del Neolític a La Península Ibèrica*. Universitat de Valencia, 7-9 d'abril 1999, SAGVNTUM 2, 59–67.

Blasco, A, Edo, M., Villalba, M. J., & Saña, M. (2003). Primeros datos sobre la utilización sepulcral de la Cueva de Can Sadurní (Begues, Baix Llobregat) en el Neolítico Cardial. In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico en la Peninsula Ibérica* (pp. 625–633). Santander: Monografías del Instituto Internacional de Investigaciones Prehistoricas de Cantabria.

Blasco, A, Edo, M., & Villalba, J. (2005). Cardial, epicardial y postcardial en Can Sadurní (Begues, Baix Llobregat). El largo fin del Neolítico Antiguo en Cataluña. In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico en la Peninsula Ibérica* (pp. 867–877). Santander: Monografías del Instituto Internacional de Investigaciones Prehistoricas de Cantabria.

Blasco, A, Edo, M., & Villalba, M. J. (2008). Evidencias de procesado y consumo de cerveza en la cueva de Can Sadurní (Begues, Barcelona) durante la prehistoria. In M. S. Hernández, J. A. Soler, & J. A. López (Eds.), *IV Congreso del Neolítico Peninsular* (Vol. 1, pp. 428–431). Alicante: Museo Arqueológico de Alicante.

Blasco, A, Edo, M., & Villalba, M. J. (2011). *La cova de can Sadurní i la prehistòria de Garraf. Recull de 30 anys d'investigació. Actes de Les Jornades Internacionals de Prehistòria 'El Garraf, 30 Anys d'Investigació Arqueològica.* Edar. Arqueología y Patrimonio.

Blech, M., & Marzoli, D. (2005). Cambios en el paisaje costero del Empordà. Las investigaciones interdisciplinarias llevadas a cabo por el Instituto Arqueológico Alemán, Madrid. *Empúries*, 54, 45–58.

Bofill, M. (2016). Les indústries macrolítiques. In Anna Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 117–123). Barcelona, Spain: Ajuntament de Barcelona.

Bofill, M., Clop, X., & Molist, M. (2008). Utillatge macrolític en els nivells neolítics de l'assentament de la Caserna de Sant Pau. *Quarhis,* 4, 46–49.

Bordas, A., Mora, L, R., & López, V. (1995). El assentamiento al aire libre del Neolítico en la Font del Ros (Berga, Bergueda). In M. Borrell, J. Bosch, & A. Ribes (Eds.), *I Congrés del Neolític a la Península Ibèrica.* (Vol. 1, pp. 397–406). Gavà/Bellaterra: Rubricatum.

Borrell, F. (2008). La industria lítica tallada del jaciment neolític de la Caserna de Sant Pau. *Quarhis,* (4), 36–45.

Borrell, F., Gómez, A., Molist, M., Tornero, C., & Vicente, O. (2011). Implantació, consolidació i desenvolupament de les societats agrícola-ramaderes al terç meridional de la Serralada Litoral catalana: la cova de Sant Llorenç (Sitges, Garraf). In A Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la prehistòria del Garraf. Recull de 30 anys d'investigació* (pp. 347–357). Edar. Arqueología y Patrimonio.

Borrell, F., & Molist, M. (2012). Aprovisionamiento y gestión del jaspe durante el neolítico antiguo en el noreste peninsular en el yacimiento de la Caserna de Sant Pau (Barcelona). *Archivos de Prehistoria Levantina,* (XXIV), 141–156.

Borrell, F., & Gibaja, J. F. (2012). The First Neolithic Communities in Northeast Iberia: Procurement, Production, and Use of Lithic Tools at the Settlement of Caserna de Sant Pau Del Camp (Barcelona, Spain). *The Journal of Island and Coastal Archaeology*, 7(3), 313–337.

Borrell, F., Gómez, A., Molist, M., Tornero, C., & Vicente, O. (2014). Les ocupacions de la Cova de Sant Llorenç (Sitges, Garraf): Noves aportacions al coneixement de la prehistòria del Garraf. *Tribuna d'Arqueologia*, 110–128.

Borrell, F., Gomez, A., Molist, M., Tornero, C., & Vicente, O. (2015). La cova de Sant Llorenç i el procés de neolitització de la zona del Garraf. In P. Izquierdo (Ed.), *Arqueologia de Sitges: I Jornada*, Palau de Maricel, 28 de febrer de 2014 (pp. 46–53). Sitges: Consorci del Patrimoni de Sitges.

Borrell, F., Molist, M., Tornero, C., & Vicente, O. (2016). La Cova de Sant Llorenç (Sitges, Barcelona). Un Exemple d'Ocupació en cova en el procés de neolitització del Garraf. In X Esteve, C. Miró, N. Molist, & G. Sabaté (Eds.), *Jornades d'Arqueologia del Penedès*, 2011 (1sto, pp. 97–107). Vilafranca del Penedès, Spain: Institut d'Estudis Penedesencs.

Bosch, A., & Aliaga, S. (1998). *El Poblat neolític de Plansallosa: L'explotació del territori dels primers agricultors-ramaders de l'Alta Garrotxa.* Museu Comarcal de la Garrotxa.

Bosch, A., & Tarrús, J. (2018). La cerámica. In Antoni Palomo, R. Piqué, & X. Terradas (Eds.), *La revolución Neolítica. La Draga, el poblado de los progidios.* (pp. 117–120). Madrid, Spain: Museo Arqueológico Regional de la Comunidad de Madrid.

Bosch, À., Chinchilla, J., Piqué, R., & Tarrús, J. (1996). Hallazgo de los primeros utensilios de madera en el poblado neolítico de la Draga (Banyoles, Girona) (The finding of the first wooden tools in the neolithic site of la Draga [Banyoles, Girona]). *Trabajos De Prehistoria*, 53(1), 147.

Bosch, À., Chinchilla, J., & Tarrús, J. (2000). *El poblat Lacustre Neolític de la Draga. Excavacions de 1990 a 1998.* Monografies del CASC. Museu d'Arqueologia de Catalunya.

Bosch, À., Chinchilla, J., & Tarrús, J. (2011). *El Poblat Lacustre del Neolític Antic de La Draga. Excavacions de 2000 a 2005.* Monografies del CASC. Barcelona: Museu d'Arqueologia de Catalunya.

Bosch, J. (1993). Cronologia Prehistòrica al curs inferior de l'Ebre. Primeres Datacions Absolutes. *Pyrenae*, (24), 53–56.

Bosch, J. (2005). *El proceso de neolitización en la región del curso inferior del Ebro.* PhD thesis. Universitat de Barcelona.

Bosch, J. (2016a). La cerámica de la Cova del Vidre (Roquetes) y el Neolítico Cardial Franco-Ibérico. In *Del neolític a l'edat del bronze en el Mediterrani occidental. Estudis en homenatge a Bernat Martí Oliver*. (Vol. TV SIP 119, pp. 109–115). Valencia, Spain.

Bosch, J. (2016b). La Cueva del Vidre Roquetes, Bajo Ebro. Asentamiento del Mesolítico y del Neolítico Antiguo en la Cordillera Costera Catalana meridional. In V. Gonçalves, M. Diniz, & A. C. Sousa (Eds.), *V Congresso do Neolítico Peninsular* (pp. 182–188). Lisboa: Centro de arqueologia da Universidade de Lisboa.

Bosch, J., & Estrada, A. (1994). El Neolític Postcardial a les Mines Prehistòriques de Gavà (Baix Llobregat). *Rubricatum* 0. Gavà: Museu de Gavà.

Bosch, J, Forcadell, A., & Villalbí, M. del M. (1991). Les estructures d'hàbitat a l'assentament del Barranc d'en Fabra. In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 121–122). Puigcerdà, Spain: Institut d'Estudis Ceretans.

Bosch, J., Forcadell, A., & Villalbí, M. del M. (1995a). El “Barrac de Fabra”: Asentamiento de inicios del IV Milenio AC. en el curso inferior del Ebro. In M. Borrell, J. Bosch, & A. Ribes (Eds.), *I Congrés del Neolític a la Península Ibèrica.* (Vol. 1, pp. 391–396). Gavà/Bellaterra: Rubricatum.

Bosch, J, Villalbí, M. del M., & Forcadell, A. (1995b). El barranc d ’en Fabra un assentament neolític a l’aire lliure. *Tribuna d’arqueologia*, 51–62.

Bosch, J, Estrada, A., & Juan-Muns, N. (1999). L’aprofitament de recursos faunístics aquatics , marins i Iitorals , durant el neolític a Gava (Baix Llobregat). *SAGVNTVM-PLAV*, Extra-2, 77–83.

Box, G. E. P. (1976). Science and Statistics. *Journal of the American Statistical Association*, 71(356), 791–799. https://doi.org/10.1080/01621459.1976.10480949

Brettell, R. C., Stern, B., Reifarth, N., & Heron, C. (2014). The “semblance of immortality”? Resinous materials and mortuary rites in Roman Britain. *Archaeometry*, 56(3), 444–459. https://doi.org/10.1111/arcm.12027

Breu, A. (in press). How to measure the invisible. A quantitative approach towards the analysis of prints by toothed shells. In *VI Jornades de Joves Investigadors en Arqueologia*. Universitat de Barcelona. Barcelona

Breu, A. (2014). *El fenomen cardial al noroest peninsular: una visió des de la perspectiva de l’estil*. Ma Dissertation. Universitat Autònoma de Barcelona.

Breu, A. (2015). *Organic residue analysis from pottery: testing for shape-content relations in the catalan Early Neolithic.* MSc Dissertation. University of Bradford.

Buck, C. E., Cavanagh, W. G., & Litton, C. D. (1996). *Bayesian approach to interpreting archaeological data*. Wiley. London.

Budja, M., Ogrinc, N., Žibrat Gašparič, A., Potočnik, D., Žigon, D., & Mlekuž, D. (2013). Transition to farming – transition to milk culture: a case study from Mala Triglavca, Slovenia. *Documenta Praehistorica*, 40(1), 97–117. https://doi.org/10.4312/dp.40.9

Burger, J., Kirchner, M., Bramanti, B., Haak, W., & Thomas, M. G. (2007). Absence of the Lactase-Persistence-Associated Allele in Early Neolithic Europeans. *Proceedings of the National Academy of Sciences*, 104(10), 3736–3741. https://doi.org/10.1073/pnas.0607187104

Buxó, R. (2016). La primera agricultura del pla de Barcelona: conreu i gestió. In A Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 91–95). Barcelona, Spain: Ajuntament de Barcelona.

Buxó, R, & Canal, D. (2008). L'agricultura i l'alimentació vegetal. *Quarhis*, 4(4), 54–56.

Buxó, R. (1990). Metodología y técnicas para la recuperación de restos vegetales (en especial referenca a semillas y frutos) en yacimientos arqueológicos. *Cahier Noir*, 5, 63.

C

Campbell, E. N., Housley, R. A., & Taylor, M. (2004). Charred food residues from Hebridean Iron Age pottery: analysis and dating. In R. A. Housley & G. Coles (Eds.), *Atlantic Connections and Adaptations: Economies, Environments and Subsistence in Lands Bordering the North Atlantic.* (pp. 65–85). Oxbow Books.

Cañabate, M. L., & Sánchez, A. (1995). Analisis de indicadores Bioquimicos Del Contenido De Recipientes Arqueológicos. *Complutum*, 6, 281–291.

Cañabate, M. L., & Sánchez, A. (1996). Análisis de indicadores bioquímicos en un ataifor almohade. *Arqueología y Territorio Medieval*, 3(3), 293–296.

Cañabate, M. L., & Sánchez, A. (1997). Identificación de grasas y ésteres de ceras en recipientes arqueológicos. *Caesaraugusta*, (73), 319–325.

Castro, P. V, Escoriza, T., Masclans, A., & Oltra, J. (2012). Discusión sociológica sobre las comunidades del Sudeste Ibérico en el Horizonte de Villena (c. 1550/1500 - 1300/1250 cal ANE). Un contexto histórico para las cerámicas del estilo de "Cogotas I." In J. A. Rodríguez & J. Fernández (Eds.), *Cogotas I: una cultura de la Edad del Bronce en la Península Ibérica: Homenaje a Ma Dolores Fernández-Posse* (pp. 657–580). Valladolid, Spain: Universidad de Valladolid.

Cavalli-Sforza, L. L., Piazza, A., Menozzi, P., & Mountain, J. (1988). Reconstruction of human evolution: bringing together genetic, archaeological, and linguistic data. *Proceedings of the National Academy of Sciences*, 85(16), 6002–6006.

Cebrià, A., Fontanals, M., Martín, P., Morales, J. I., Oms, F. X., Rodríguez-Hidalgo, A., ... Vergès, J. M. . (2014). Nuevos datos para el Neolítico antiguo en el nordeste de la Península Ibérica procedentes de la Cova del Toll (Moià, Barcelona) y de la Cova de la Font Major (L'Espluga de Francolí, Tarragona). *Trabajos de Prehistoria*, 71(1), 134–145. https://doi.org/10.3989/tp.2014.12128

Charters, S., Evershed, R. P., Goad, L. J., Heron, C., & Blinkhorn, P. W. (1993a). Identification Of An Adhesive Used To Repair A Roman Jar. *Archaeometry*, 35(1), 91–101. https://doi.org/10.1111/j.1475-4754.1993.tb01025.x

Charters, S., Evershed, R. P., Goad, L. J., Leyden, A., Blinkhorn, P. W., & Denham, V. (1993b). Quantification and distribution of lipid in archaeological ceramics: implications for sampling potsherds for organic residue analysis and the classification of vessel use. *Archaeometry*, 35(2), 211–223. https://doi.org/10.1111/j.1475-4754.1993.tb01036.x

Charters, S., Evershed, R. P., Blinkhorn, P. W., & Denham, V. (1995). Evidence for the mixing of fats and waxes in archaeological ceramics. *Archaeometry*, 37(1), 113–127. https://doi.org/10.1111/j.1475-4754.1995.tb00730.x

Charters, S., Evershed, R. P., Quye, A., Blinkhorn, P. W., & Reeves, V. (1997). Simulation Experiments for Determining the Use of Ancient Pootery Vessels: the Behaviour of Epicuticular Leaf Wax During Boiling of a Leafy Vegetable. *Journal of Archaeological Science*, 24(1), 1–7. https://doi.org/http://dx.doi.org/10.1006/jasc.1995.0091.

Clausell, G., Izquierdo, I., Arasa, F., & Tresserras, J. (2000). La fase del ibérico final en el asentamiento del Torrelló del Boverot (Alzamora, Castellón): dos piezas cerámicas singulares. *Archivo Español de Arqueología*, 73, 87–104.

Clop, X. (2002). Producción de cerámicas y funcionalidad durante el IIIer milenio cal ane en el noreste de la península Ibérica. In *Análisis funcional: su aplicación al estudio de sociedades prehistóricas* (pp. 251–260). Archaopress.

Clop, X. (2011). Caracterización petroarqueológica de cerámicas decoradas del Neolítico Antiguo de la Peninsula Ibérica. In J. Bernabeu, M. Rojo, & L. Molina (Eds.), *Las primeras producciones cerámicas: el VI milenio cal aC en la Península Ibérica.* (pp. 35–52).

Clop, X, Faura, J. M., & Gangonells, M. (1992). La cronologia absoluta del Neolític Antic a Catalunya: una visió de conjunt. In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 63–65). Puigcerdà, Spain: Institut d'Estudis Ceretans.

Clop, X., Saña, M., & Terradas, X. (2008). Algunes consideracions entorn al "temps" en el mesolític, neolític i calcolític de Catalunya. *Cypsela*, 17, 35–50.

Colombini, M. P., Giachi, G., Modugno, F., & Ribechini, E. (2005). Characterisation of organic residues in pottery vessels of the Roman age from Antinoe (Egypt). *Microchemical Journal*, 79(1–2), 83–90. https://doi.org/10.1016/j.microc.2004.05.004

Colominas, J. (1925). *Prehistòria de Montserrat.* Montserrat: Monestir de Montserrat.

Colominas, L., Lladó, R., Saña, M., & Tornero, C. (2008). La gestió dels recursos animals durant les ocupacions de l'assentament de la caserna de Sant Pau. *Quarhis*, 4(4), 57–63.

Colonese, A. C., Farrell, T., Lucquin, A., Firth, D., Charlton, S., Robson, H. K., ... Craig, O. E. (2015). Archaeological bone lipids as palaeodietary markers. *Rapid Communications in Mass Spectrometry: RCM,* 29(7), 611–618. https://doi.org/10.1002/rcm.7144

Colonese, A. C., Hendy, J., Lucquin, A., Speller, C. F., Collins, M. J., Carrer, F., ... Craig, O. E. (2017a). New criteria for the molecular identification of cereal grains associated with archaeological artefacts. *Scientific Reports*, 7(1), 6633. https://doi.org/10.1038/s41598-017-06390-x

Colonese, A. C., Lucquin, A., Guedes, E. P., Thomas, R., Best, J., Fothergill, B. T., ... Craig, O. E. (2017b). The identification of poultry processing in archaeological ceramic vessels using in-situ isotope references for organic residue analysis. *Journal of Archaeological Science*, 78, 179–192. https://doi.org/10.1016/j.jas.2016.12.006

Condamin, J., Formenti, F., Metais, M. O., Michel, M., & Blond, P. (1976). The Application Of Gas Chromatography To The Tracing Of Oil In Ancient Amphorae. *Archaeometry*, 18(2), 195–201. https://doi.org/10.1111/j.1475-4754.1976.tb00160.x

Connan, J., & Van de Velde, T. (2010). An overview of bitumen trade in the Near East from the Neolithic (c.8000 BC) to the early Islamic period. *Arabian Archaeology and Epigraphy*, 21(1), 1–19. https://doi.org/10.1111/j.1600-0471.2009.00321.x

Cook, D. A., Zbitnew, A., Dempster, G., & Gerrard, J. W. (1978). Detection of antibody to rotavirus by counterimmunoelectrophoresis in human serum, colostrum and milk. *Journal of Pediatrics*, 93(6), 967–970.

Copley, M. S., Rose, P. J., Clapham, A., Edwards, D. N., Horton, M. C., & Evershed, R. P. (2001). Detection of palm fruit lipids in archaeological pottery from Qasr Ibrim, Egyptian Nubia. *Proceedings of the Royal Society B: Biological Sciences*, 268(1467), 593–597. https://doi.org/10.1098/rspb.2000.1394

Copley, M. S., Berstan, R., Dudd, S. N., Docherty, G., Mukherjee, A. J., Straker, V., ... Evershed, R. P. (2003). Direct Chemical Evidence for Widespread Dairying in Prehistoric Britain. *Proceedings of the National Academy of Sciences*, 100(4), 1524–1529. https://doi.org/10.1073/pnas.0335955100

Copley, M. S., Hansel, F. A., Sadr, K., & Evershed, R. P. (2004). Organic residue evidence for the processing of marine animal products in pottery vessels from the pre-colonial archaeological site of Kasteelberg D east, South Africa. *South African Journal of Science*, 100, 279–283.

Copley, M. S., Berstan, R., Dudd, S. N., Aillaud, S., Mukherjee, A. J., Straker, V., ... Evershed, R. P. (2005a). Processing of milk products in pottery vessels through British prehistory. *Antiquity*, 79(306), 895–908.

Copley, M. S., Berstan, R., Dudd, S. N., Straker, V., Payne, S., & Evershed, R. P. (2005b). Dairying in antiquity. I. Evidence from absorbed lipid residues dating to the British Iron Age. *Journal of Archaeological Science*, 32(4), 485–503. https://doi.org/10.1016/j.jas.2004.07.004

Copley, M. S., Berstan, R., Dudd, S. N., Straker, V., Payne, S., & Evershed, R. P. (2005c). Dairying in antiquity. II. Evidence from absorbed lipid residues dating to the British Bronze Age. *Journal of Archaeological Science*, 32(4), 505–521. https://doi.org/10.1016/j.jas.2004.07.004

Copley, M. S., Berstan, R., Mukherjee, A. J., Dudd, S. N., Straker, V., Payne, S., & Evershed, R. P. (2005d). Dairying in antiquity. III. Evidence from absorbed lipid residues dating to the British Neolithic. *Journal of Archaeological Science*, 32(4), 523–546. https://doi.org/10.1016/j.jas.2004.08.006

Copley, M. S., Bland, H. A., Rose, P., Horton, M. C. M., & Evershed, R. P. (2005e). Gas chromatographic, mass spectrometric and stable carbon isotopic investigations of organic residues of plant oils and animal fats employed as illuminants in archaeological lamps from Egypt. *Analyst*, 130(6), 860–871. https://doi.org/10.1039/B500403A

Correa-Ascencio, M., & Evershed, R. P. (2014). High throughput screening of organic residues in archaeological potsherds using direct acidified methanol extraction. *Analytical Methods*, 6(5), 1330–1340. https://doi.org/10.1039/C3AY41678J

Correa-Ascencio, M., Robertson, I. G., Cabrera-Cortés, O., Cabrera-Castro, R., & Evershed, R. P. (2014). Pulque production from fermented agave sap as a dietary supplement in Prehispanic Mesoamerica. *Proceedings of the National Academy of Sciences*, 111(39), 14223–14228. https://doi.org/10.1073/pnas.1408339111

Costa, F., García, P., Marcet, R., & Mas, J. (1982). El jaciment prehistòric de Can Soldevila (Santa Perpètua de Mogoda). In Grup pro-arqueologia i història (Ed.), *Fulls d'arqueologia i història de Santa Perpètua de Mogoda* (pp. 9–48). Santa Perpètua de Mogoda: Grup pro-arqueologia i història.

Courel, B. (2016). *Caractérisation de substances naturelles en contexte archéologique: apport des études moléculaires, isotopiques et de la datation au ^{14}C*. PhD Thesis. Institut de Chimie. Université de Strasbourg.

Craig, O. E. (2002). The Development of Dairying in Europe: potential evidence from food residues on ceramics. *Documenta Praehistorica*, 29, 97–107. Retrieved from http://eprints.whiterose.ac.uk/54355/

Craig, O. E. (2004). Organic analysis of "food Crusts" from sites in the Schelde valley, Belgium: a preliminary evaluation. *Notae Praehistoricae*, 24, 209–217.

Craig, O. E. (2006). El desenvolupament de la producció de llet a Europa: l'evidència a partir dels residus d'aliments a les ceràmiques. *Cota Zero*, 21, 74–86.

Craig, O. E., Chapman, J., Heron, C., Willis, L. H., Bartosiewicz, L., Taylor, G., ... Collins, M. J. (2005a). Did the First Farmers of Central and Eastern Europe Produce Dairy Foods? *Antiquity*, 79, 882–894.

Craig, O. E., Taylor, G., Mulville, J., Collins, M. J., & Parker, M. (2005b). The identification of prehistoric dairying activities in the Western Isles of Scotland: an integrated biomolecular approach. *Journal of Archaeological Science*, 32(1), 91–103. https://doi.org/http://dx.doi.org/10.1016/j.jas.2004.06.009

Craig, O. E., Forster, M. D., Andersen, S. H., Koch, E., Crombé, P., Milner, N. J., ... Heron, C. (2007). Molecular and isotopic demonstration of the processing of aquatic products in northern european prehistoric pottery. *Archaeometry*, 49(1), 135–152. https://doi.org/10.1111/j.1475-4754.2007.00292.x

Craig, O. E., Steele, V. J., Fischer, A., Hartz, S., Andersen, S. H., Donohoe, P., ... Heron, C. (2011). Ancient lipids reveal continuity in culinary practices across the transition to agriculture in Northern Europe. *Proceedings of the National Academy of Sciences*, 108(44), 17910–17915. https://doi.org/10.1073/pnas.1107202108

Craig, O. E., Allen, R. B., Thompson, A., Stevens, R. E., Steele, V. J., & Heron, C. (2012). Distinguishing wild ruminant lipids by gas chromatography/combustion/isotope ratio mass spectrometry. *Rapid Communications in Mass Spectrometry*, 26(19), 2359–2364. https://doi.org/10.1002/rcm.6349

Craig, O. E., Saul, H., Lucquin, A., Nishida, Y., Taché, K., Clarke, L., ... Jordan, P. (2013). Earliest evidence for the use of pottery. *Nature*, 496(7445), 351–354. https://doi.org/10.1038/nature12109

Craig, O. E., Shillito, L.-M., Albarella, U., Viner-Daniels, S., Chan, B., Cleal, R., ... Pearson, M. P. (2015). Feeding Stonehenge: cuisine and consumption at the Late Neolithic site of Durrington Walls. *Antiquity*, 89(347), 1096–1109. https://doi.org/10.15184/aqy.2015.110

Cramp, L. J. E., & Evershed, R. P. (2014). Reconstructing Aquatic Resource Exploitation in Human Prehistory Using Lipid Biomarkers and Stable Isotopes. In *Treatise on Geochemistry* (Vol. 14, pp. 319–339). Elsevier. https://doi.org/10.1016/B978-0-08-095975-7.01225-0

Cramp, L. J. E., Evershed, R. P., & Eckardt, H. (2011). What was a mortarium used for? Organic residues and cultural change in Iron Age and Roman Britain. *Antiquity*, 85(330), 1339–1352. https://doi.org/doi:10.1017/S0003598X00062098

Cramp, L. J. E., Evershed, R. P., Lavento, M., Halinen, P., Mannermaa, K., Oinonen, M., ... Heyd, V. (2014a). Neolithic dairy farming at the extreme of agriculture in northern Europe. *Proceedings of the Royal Society B: Biological Sciences*, 281(1791), 20140819. https://doi.org/10.1098/rspb.2014.0819

Cramp, L. J. E., Jones, J., Sheridan, A., Smyth, J., Whelton, H. L., Mulville, J., ... Evershed, R. P. (2014b). Immediate replacement of fishing with dairying by the earliest farmers of the Northeast Atlantic archipelagos. *Proceedings of the Royal Society B: Biological Sciences*, 281(1780), 20132372. https://doi.org/10.1098/rspb.2013.2372

Cramp, L. J. E., Ethier, J., Urem-Kotsou, D., Bonsall, C., Borić, D., Boroneanţ, A., ... Ivanova, M. (2019). Regional diversity in subsistence among early farmers in Southeast Europe revealed by archaeological organic residues. *Proceedings of the Royal Society B: Biological Sciences*, 286(1894), 20182347. https://doi.org/10.1098/rspb.2018.2347

Cruz, I., & Ortega, N. (2011). Integración en el discurso estratigráfico actual de los materiales prehistóricos procedentes de excavaciones antiguas de la cueva de Can Sadurní, ubicados en la Biblioteca-Museo Víctor Balaguer de Vilanova i la Geltrú. In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació*. (pp. 159–163). Edar. Arqueología y Patrimonio.

Cura, M., Ramis, F., & Padró, J. (1979). En torno a un hallazgo del Neolítico Antiguo en la Valldany (Oliana, comarca del Urgellet, Lérida). *Pyrenae*: Revista de Prehistòria i Antiguitat de La Mediterrània Occidental, 1979, Vol. 15-16, p. 371-376.

D

Daura, J., Sanz, M., Oms, F. X., Pedro, M., Martínez, P., Mendiela, S., ... Fullola, J. M. (2019). Deciphering Neolithic activities from a Cardial burial site (Cova Bonica) on the western Mediterranean coast. *Journal of Archaeological Science: Reports*, 23(October 2018), 324–347. https://doi.org/10.1016/j.jasrep.2018.10.036

de Beaune, S. A. (1987). Palaeolithic Lamps and Their Specialization: A Hypothesis. *Current Anthropology*, 28(4), 569–577.

De la Vega, J. (1972). Materiales arqueológicos de la cova Cassimanya (Begues). *Mediterrània*, 7, 1–7.

Dedet, B., & Py, M. (1975). *Classification de la céramique non tournée protohistorique du Languedoc méditerranéen* (Vol. 4). Paris: Diffusion de Boccard.

DeNiro, M. J., & Epstein, S. (1977). Mechanism of carbon isotope fractionation associated with lipid synthesis. *Science*, 197(4300), 261–263.

DeNiro, M. J., & Epstein, S. (1978). Influence of diet on the distribution of carbon isotopes in animals. *Geochimica et Cosmochimica Acta*, 42(5), 495–506.

Desse, J. (1987). La pêche: son rôle dans l'économie des premières sociétés néolithiques en Méditerranée occidentale. In J. Guilaine, (Ed.). *Premières communautés paysannes en Méditerranée occidentale: Actes du Colloque International du CNRS* (Montpellier, 26-29 avril 1983). (pp. 281–285). Centre national de la recherche scientifique.

Dobson, G., Christie, W. W., & Sebedio, J. L. (1996a). Monocyclic saturated fatty acids formed from oleic acid in heated sunflower oils. Chemistry and *Physics of Lipids*, 82, 101–110.

Dobson, G., Christie, W. W., & Sebedio, J. L. (1996b). The nature of cyclic fatty acids formed in heated vegetable oils. *Grasas y Aceites*, 47(1–2), 26–33.

Dobson, G., & Sebedio, J. L. (1999). Monocyclic dienoic fatty acids formed from γ -linolenic acid in heated evening primrose oil. *Chemistry and Physics of Lipids*, 97, 105–118.

Drieu, L, Horgines, M., Binder, D., Pétrequin, P., Pétrequin, A.-M., Peche-Quilichini, K., … Regert, M. (2019). Influence of Porosity on Lipid Preservation in the Wall of Archaeological Pottery. *Archaeometry*, (April). https://doi.org/10.1111/arcm.12479

Drieu, L. (2017). *Fabrication et usages des poteries durant le Néolithique et la Protohistoire en Europe : Les apports de l'archéologie biomoléculaire*. PhD Thesis.Université Côte d'Azur.

Dudd, S. N. (1999). *Molecular and Isotopic Characterisation of Animal Fats in Archaeological Pottery*. PhD Thesis. Bristol University.

Dudd, S. N., & Evershed, R. P. (1998). Direct Demonstration of Milk as an Element of Archaeological Economies. *Science*, 282(5393), 1478–1481. https://doi.org/10.1126/science.282.5393.1478

Dudd, S. N., Regert, M., & Evershed, R. P. (1998). Assessing microbial lipid contributions during laboratory degradations of fats and oils and pure triacylglycerols absorbed in ceramic potsherds. *Organic Geochemistry*, 29(5–7), 1345–1354.

Dudd, S. N., Evershed, R. P., & Gibson, A. M. (1999). Evidence for Varying Patterns of Exploitation of Animal Products in Different Prehistoric Pottery Traditions Based on Lipids Preserved in Surface and Absorbed Residues. *Journal of Archaeological Science*, 26(12), 1473–1482. https://doi.org/10.1006/jasc.1998.0434

Dunne, J., Evershed, R. P., Salque, M., Cramp, L., Bruni, S., Ryan, K., … Di Lernia, S. (2012). First dairying in green Saharan Africa in the fifth millennium bc. *Nature*, 486(7403), 390–394. https://doi.org/10.1038/nature11186

Dunne, J., Mercuri, A. M., Evershed, R. P., Bruni, S., & di Lernia, S. (2016). Earliest direct evidence of plant processing in prehistoric Saharan pottery. *Nature Plants*, 3(December), 16194. https://doi.org/10.1038/nplants.2016.194

E

Edo, M., Millán, M., Blasco, A., & Blanch, M. (1986). Resultats de les excavacions de la Cova de Can Sadurní (Begues, Baix Llobregat). *Tribuna d'Arqueologia*, 1985–1986, 33–41.

Edo, M., Blasco, A., & Villalba, M. J. (2011). La cova de Can Sadurní, guió sintètic de la prehistòria recent de Garraf. In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació*. (pp. 13–97). Edar. Arqueología y Patrimonio.

Edo, M., Ferran, A., Bardera, R., Saña, M., Bergadà, M. M., Barrio, M. J., ... Fornell, E. (2017). Cova de Can Sadurní, la transformació d'un jaciment. L'episodi sepulcral del neolític postcardial. *Tribuna d'arqueologia*. 2013, 81-105.

Edo, M., Antolín, F., Martínez-Rodríguez, P., Villalba, M. J., Fullola, J. M., Bergadà, M. M., ... Gibaja, J. F. (2019). La cueva de Can Sadurní (Begues, Barcelona): el episodio funerario del neolítico antiguo cardial pleno. Estado actual de la cuestión. In J. F. Gibaja, M. Mozota, M. E. Subirà, A. Martín, & J. Roig (Eds.), *Mirando a la muerte. Las prácticas funerarias durante el neolítico en el noreste peninsular*. Vol. 3 (pp. 205–304). Castellón de la Plana: e-DitARX.

Equip Guineu. (1994). Elaboració d'una cronoestratigrafia per a la prehistòria del Penedès. *Tribuna d'arqueologia*, 7–24.

Esteve, F. (2000). *Recerques Arqueològiques a la Ribera Baixa de l'Ebre*. I. Prehistòria. Amposta. Museu del Montsià. Ajuntament d'Amposta.

Esteve, X., Martín, P., Oms, F. X., López, D., & Jornet, R. (2010). *Intervencions arqueològiques als enllaços de l 'autopista AP-7 de Vilafranca del Penedès : nous assentaments prehistòrics a l 'aire lliure al Penedès.* Unpublished report.

Ethier, J., Bánffy, E., Vuković, J., Leshtakov, K., Bacvarov, K., Roffet-Salque, M., ... Ivanova, M. (2017). Earliest expansion of animal husbandry beyond the Mediterranean zone in the sixth millennium BC. *Scientific Reports*, 7(1), 7146. https://doi.org/10.1038/s41598-017-07427-x

Evershed, R. P. (1993). Biomolecular Archaeology and Lipids. *World Archaeology*, 25(1), 74–93. https://doi.org/10.2307/124755

Evershed, R. P. (1999). Lipids as carriers of anthropogenic signals from prehistory. *Philosophical Transactions of the Royal Society B: Biological Sciences,* 354(1379), 19–31. https://doi.org/10.1098/rstb.1999.0357

Evershed, R. P. (2008). Organic residue analysis in archaeology: the archaeological biomarker revolution. *Archaeometry*, 50(6), 895–924. https://doi.org/10.1111/j.1475-4754.2008.00446.x

Evershed, R. P., & Connolly, R. C. (1988). Lipid preservation in Lindow Man. *Naturwissenschaften*, 75, 143–145.

Evershed, R.P., & Charters, S. (1995). Simulating the degradation of animal fats in archaeological ceramics. In J. Grimalt & C. Dorronsoro (Eds.), *Organic Geochemistry: developments and applications to energy, climate, environment and human history*. (pp. 697–698). Donostia: European Association of Organic Geochemists.

Evershed, R. P., Jerman, K., & Eglinton, G. (1985). Pine wood origin for pitch from the Mary Rose. *Nature*, 314, 525–528.

Evershed, R. P., Heron, C., & Goad, L. J. (1990). Analysis of organic residues of archaeological origin by high-temperature gas chromatography and gas chromatography-mass spectrometry. *Analyst*, 115(10), 1339–1342. https://doi.org/10.1039/AN9901501339

Evershed, R. P., Heron, C., Charters, S., & Goad, L. J. (1991a). The Survival of Food Residues: New Methods of Analysis, Interpretation and Application. *Proceedings of the British Academy*, 77(February 1991), 187–208.

Evershed, R P, Heron, C., & Goad, L. J. (1991b). Epicuticular wax components preserved in potsherds as chemical indicators of leafy vegetables in ancient diets. *Antiquity*, 65(248), 540–544. https://doi.org/doi:10.1017/S0003598X00080145

Evershed, R P, Heron, C., Charters, S., & Goad, L. J. (1992). The Survival Of Food Residues: New Methdos Of Analysis, Interpretation And Application. *Proceedings of the British Academy*, 77, 187–208.

Evershed, R P, Arnot, K. I., Collister, J., Eglinton, G., & Charters, S. (1994). Application of isotope ratio monitoring gas chromatography-mass spectrometry to the analysis of organic residues of archaeological origin. *Analyst*, 119(5), 909–914. https://doi.org/10.1039/AN9941900909

Evershed, R. P., Stott, A. W., Raven, A. M., Dudd, S. N., Charters, S., & Leyden, A. (1995a). Formation of long-chain ketones in ancient pottery vessels by pyrolysis of acyl lipids. *Tetrahedron Letters*, 36(48), 8875–8878. https://doi.org/10.1016/0040-4039(95)01844-8

Evershed, R. P., Charters, S., & Quye, A. (1995b). Interpreting lipid residues in archaeological ceramics: preliminary results from laboratory simulations of vessel use and burial. *Materials Research Society Symposium Proceedings,* 352, 85–95.

Evershed, R. P., Mottram, H. R., Dudd, S. N., Charters, S., Stott, A. W., Lawrence, G. J., ... Reeves, V. (1997a). New Criteria for the Identification of Animal Fats Preserved in Archaeological Pottery. *Naturwissenschaften*, 84(9), 402–406. https://doi.org/10.1007/s001140050417

Evershed, R. P., Vaughan, S. J., Dudd, S. N., & Soles, J. S. (1997b). Fuel for thought ? Beeswax in lamps and conical cups from Late Minoan Crete. *Antiquity*, 71(1997), 979–985. https://doi.org/10.1017/S0003598X00085860

Evershed, R. P., van Bergen, P. F., Peakman, T. M., Leigh-Firbank, E. C., Horton, M. C., Edwards, D., ... Rowley-Conwy, P. A. (1997c). Archaeological frankincense. *Nature*, 390(6661), 667–668. https://doi.org/10.1038/37741

Evershed, R. P., Dudd, S. N., Copley, M. S., Berstan, R., Stott, A. W., Mottram, H. R., ... Crossman, Z. (2002a). Chemistry of Archaeological Animal Fats. *Accounts of Chemical Research,* 35(8), 660–668. https://doi.org/10.1021/ar000200f

Evershed, R. P., Dudd, S. N., Copley, M. S., & Mutherjee, A. (2002b). Identification of animal fats via compound specific $\delta^{13}C$ values of individual fatty acids: assessments of results for reference fats and lipid extracts of archaeological pottery vessels. *Documenta Praehistorica*, 21, 73–96.

Evershed, R. P., Dudd, S. N., Anderson-Stojanovic, V. R., & Gebhard, E. R. (2003). New Chemical Evidence for the Use of Combed Ware Pottery Vessels as Beehives in Ancient Greece. *Journal of Archaeological Science*, 30(1), 1–12. https://doi.org/http://dx.doi.org/10.1006/jasc.2001.0827

Evershed, R. P., Copley, M. S., Dickson, L., & Hansel, F. A. (2008a). Experimental Evidence For The Processing Of Marine Animal Products And Other Commodities Containing Polyunsaturated Fatty Acids In Pottery Vessels. *Archaeometry*, 50(1), 101–113. https://doi.org/10.1111/j.1475-4754.2007.00368.x

Evershed, Richard P, Payne, S., Sherratt, A. G., Copley, M. S., Coolidge, J., Urem-Kotsu, D., ... Burton, M. M. (2008b). Earliest date for milk use in the Near East and southeastern Europe linked to cattle herding. *Nature*, 455(7212), 528–531. https://doi.org/10.1038/nature07180

F

Fernandes, R., Eley, Y., Brabec, M., Lucquin, A., Millard, A., & Craig, O. E. (2017). Reconstruction of prehistoric pottery use from fatty acid carbon isotope signatures using Bayesian inference. *Organic Geochemistry*, 117, 31–42. https://doi.org/10.1016/j.orggeochem.2017.11.014

Fernandes, R., Grootes, P., Nadeau, M.-J., & Nehlich, O. (2015). Quantitative diet reconstruction of a Neolithic population using a Bayesian mixing model (FRUITS): The case study of Ostorf (Germany). *American Journal of Physical Anthropology*, 158(2), 325–340. https://doi.org/10.1002/ajpa.22788

Fernandes, R., Millard, A. R., Brabec, M., Nadeau, M. J., & Grootes, P. (2014). Food reconstruction using isotopic transferred signals (FRUITS): A bayesian model for diet reconstruction. *PLoS ONE*, 9(2), 1–9. https://doi.org/10.1371/journal.pone.0087436

Fíguls, A. (1995). Estudi de l'estratègia econòmica de la comunitat del neolític antic, establerta al Pla de la Bruguera (Castellar del Vallès, Vallès Occidental). *Limes*, 4(5), 19–28.

Flatz, G., & Rotthauwe, H. (1973). Lactose nutrition and natural selection. *The Lancet*, 302(7820), 76–77.

Flors, E., & Sanfeliu, D. (2011). La Cerámica Neolítica de Costamar (Cabanes, Castellón). *SAGVNTUM*. Extra-12, 187–201.

Fontanals, M. (2005). *Memòria de l'excavació preventiva realitzada al Jaciment de el Cavet (Cambrils, Baix Camp).* Unpublished report.

Fontanals, M., Euba, I., Morales, J. I., Oms, F. X., & Vergés, J. M. (2008a). El asentamiento litoral al aire libre de El Cavet (Cambrils, Tarragona). In M. S. Hernández, J. A. Soler, & J. A. López (Eds.), *IV Congreso del Neolítico Peninsular* (Vol. 1, pp. 168–175). Alicante: Museo Arqueológico de Alicante.

Fontanals, M., Belén, A., Morales, J. I., Vergès, J. M., Itzaso, E., & Oms, F. X. (2008b). L'assentament litoral a l'aire lliure del Cavet (Cambrils, Tarragona). *Butlletí Arqueològic.* Reial Societat Arqueològica Tarraconense, 30, 5–28. https://www.raco.cat/index.php/ButlletiArq/article/view/314904/405053.

Fortó, A., Martínez Rodríguez, P., & Muñoz Rufo, V. (2005). El jaciment arqueològic de Ca l'Estrada (Canovelles). *Lauro*, 28, 5–16.

Fox, A., Heron, C., & Sutton, M. Q. (1995). Characterization Of Natural Products On Native American Archaeological And Ethnographic Materials From The Great Basin Region, U.S.A.: A Preliminary Study. *Archaeometry*, 37(2), 363–375. https://doi.org/10.1111/j.1475-4754.1995.tb00749.x

Francis, G. W. (1981). *For the determination of double-bond position in unsaturated fatty acid esters.* Department of Chemistry, University of Bergen, 5000 Bergen (Norway), 29, 369–374.

Fugazzola-Delpino, M. A. (2002). Le ceramiche impresse nel Neolitico antico: Italia e Mediterraneo. Instituto Poligrafico e Zecca dello Stato.

G

Gamba, C., Jones, E. R., Teasdale, M. D., McLaughlin, R. L., Gonzalez-Fortes, G., Mattiangeli, V., ... Pinhasi, R. (2014). Genome flux and stasis in a five millennium transect of European prehistory. *Nature Communications*, 5. https://doi.org/10.1038/ncomms6257.

Gámez, D., Angelet, C., Torrades, P., & Llorens, F. (2011). Evolució de la línea de costa del Delta del Llobregat durant l'Holocè (Epipaleolític-actualitat). In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació.* (pp. 187–191). Edar. Arqueología y Patrimonio.

García-Roselló, J. (2007). La Etnoarqueología como experimentación. Identificación de marcas de manufactura en cerámicas modeladas a mano. In M. Ramos, J. E. González-Urquijo, & J. Baena (Eds.), *Arqueología experimental en la Península Ibérica: investigación, didáctica y patrimonio* (pp. 45–57). Santander: Asociación Española de Arqueología Experimental.

García, A. (2013). *La Química en Arqueología: Una Aproximación al Análisis de Contenidos.* JIA 2013. Unpublished report.

García, R. (2010). Contenidos inorgánicos y orgánicos de Ratinhos. Análisis mineralógicos y químicos. In L. Berrocal-Rangel & A. Silva (Eds.), O Castro dos Ratinhos (Barragem do Alqueva, Moura). *Escavaçoes num povoado proto histórico do Guadiana. 2004-2006* (pp. 402–408). Lisboa: Museu Nacional de Arqueologia.

García, P., Marcet, R., & Mas, J. (1982). Can Banús, estructura a l'aire lliure del neolític antic. In Grup pro-arqueologia i història (Eds.), F*ulls d'arqueologia i història de Santa Perpètua de Mogoda* (pp. 61–70). Santa Perpètua de Mogoda.

Garnier, N., Rolando, C., Høtje, J. M., & Tokarski, C. (2009). Analysis of archaeological triacylglycerols by high resolution nanoESI, FT-ICR MS and IRMPD MS/MS: Application to 5th century BC-4th century AD oil lamps from Olbia (Ukraine). *International Journal of Mass Spectrometry*, 284(1–3), 47–56. https://doi.org/10.1016/j.ijms.2009.03.003

Garnier, N., & Valamoti, S. M. (2016). Prehistoric wine-making at Dikili Tash (Northern Greece): Integrating residue analysis and archaeobotany. *Journal of Archaeological Science*, 74, 195–206. https://doi.org/10.1016/j.jas.2016.03.003

Gassiot Ballbè, E., Mazzucco, N., Obea Gómez, L., Tarifa Mateo, N., Antolín, F., Clop i Garcia, X., ... Saña Seguí, M. (2015). La Cova del Sardo de Boí i l'explotació de l'alta muntanya als Pirineus occidentals en època neolítica. *Tribuna d'arqueologia,* 2012, 196–215.

Gassiot, E., Rodríguez-Antón, D., Burjachs, F., Antolín, F., & Ballesteros, A. (2012). Poblamiento, explotación y entorno natural de los estadios alpinos y subalpinos del Pirineo central durante la primera mitad del Holoceno. *Cuaternario y Geomorfología*, 26(3–4), 29–45.

Genera, M., Galindo, A., Vílchez, J. L., Manzano, E., Blanc, M. R., Cantanrero, A., … Fernandes, G. (2015). Nuevos datos arqueométricos acerca de la cerámica del asentamiento protohistórico del Puig Roig del Roget, Masroig, Priorat: estudio mineralógico, análisis textural y de los residuos orgánicos. In O. Mercadal (Ed.), *XV Coloquio Internacional de Arqueología de Puigcerdà*. Puigcerdà: Institut d'Estudis Ceretans.

Gibaja, F. J. (2008). La funció de l'utillatge lític tallat documentat al jaciment neolític de la caserna de Sant Pau. *Quarhis*, 4(4), 46–47.

Gibaja, F. J., Palomo, A., Blasco, A., Villalba, M. J., & Edo, M. (2011). El utillaje lítico del neolítico antiguo de Can Sadurní: primeros datos. In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació*. (pp. 121–128). Edar. Arqueología y Patrimonio.

Gibaja, F. J., Oms, F. X., Mestres, J., Mazzucco, N., & Palomo, A. (2018a). Primeros resultados sobre la función del utillaje lítico de las primeras comunidades neolíticas asentadas en Les Guixeres de Vilobí (Sant Martí Sarroca, Barcelona). *SAGVNTVM*, 50, 35–56.

Gibaja, F. J., Morell, B., López-Onaindia, D., Zemour, A., Bosch, À., Tarrús, J., & Subirà, M. E. (2018b). Nuevos datos cronológicos sobre la cueva sepulcral neolítica de l'Avellaner (Les Planes d'Hostoles, Girona). *Munibe Antropologia-Arkeologia*, (7), 145–156. https://doi.org/10.21630/maa.2018.69.01

Gillis, R., Carrère, I., Saña Seguí, M., Radi, G., & Vigne, J.-D. (2016). Neonatal Mortality, Young Calf Slaughter and Milk Production during the Early Neolithic of North Western Mediterranean. *International Journal of Osteoarchaeology*, 26(2), 303–313. https://doi.org/10.1002/oa.2422

Gómez, A. (2016). Les produccions ceràmiques. In A. Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 125–135). Barcelona, Spain: Ajuntament de Barcelona.

Gómez, A. (2019). Notes a la ceràmica prehistòrica. *Anuari d'Arqueologia i Patrimoni de Barcelona*, 2017, 37.

Gómez, A., & Molist, M. (2016). Les primeres ocupacions estables al pla de Barcelona, del VI al IV mil·lenni: el desenvolupament dels agricultors-ramaders. In M. Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 35–49). Barcelona, Spain: Ajuntament de Barcelona.

Gómez, A., & Molist, M. (2017). Caserna de Sant Pau del Camp (Barcelona): Noves dades per al Neolític Antic Cardial del Nord-Est peninsular. *Cypsela*, (20), 11–23.

Gómez, A., Guerrero, E., Clop, X., Bosch, J., & Molist, M. (2008). Estudi de la ceràmica neolítica del jaciment de Caserna de Sant Pau. *Quarhis*, 4, 25–35.

Gómez, A., Alberch, P., Calvo, S., Clop, X., & Molist, M. (2013). La gestión del producto cerámico en el yacimiento de Caserna de Sant Pau del Camp (Barcelona) en la Edad del Bronce. In L. Girón, M. Lazarich, & M. Conceiçâo (Eds.), *Actas del I Primer Congreso Internacional subre Estudios Cerámicos* (pp. 452–476). Cádiz: Universidad de Cádiz. Servicio de Publicaciones.

Gómez, A., Alberch, P., Breu, A., & Molist, M. (2014a). Aproximació al gest tecnològic: La decoració cardial de Caserna de Sant Pau del Camp (Barcelona). In A Palomo, R. Piqué, & X. Terradas (Eds.), *Experimentación en Arqueología. Estudio y difusión del pasado* (pp. 363–371). Girona: Museu d'Arqueologia de Catalunya.

Gómez, A., Nadal, J., & Molist, M. (2014b). El mar i les comunitats humanes al llarg de la prehistòria al Pla de Barcelona. *Barcelona Quaderns d'Història*, 21, 63–76.

Gónzalez, J., & Harzbecher, K. (2008). Carrer de la Reina Amàlia 31-33, Carrer de la Lleialtat 1-9, i Carrer de les Carreteres 58-46. *Anuari d'Arqueologia i Patrimoni de Barcelona*, 58–60.

González, J., Harzbecher, K., & Molist, M. (2011). Un nou assentament del V mil· lenni a la costa de Barcelona. *Quarhis*, 7, 86–100.

González, J., Breu, A., Gómez Bach, A., & Molist, M. (2017). The dynamics of use and amortisation of epicardial hut at the Reina Amalia 31-33 Site (Barcelona). In *CEUR Workshop Proceedings* (Vol. 2024).

Gràcia, J. (1987). Els materials ceràmics del Neolític Cardial de la Pedrera de Sant Jordi (Pacs, Alt Penedès, Barcelona). *Miscel·lània Penedesenca*, 10, 7–22.

Granados, O., Puig, F., & Farré, R. (1993). La intervenció arqueològica a Sant Pau del Camp: un nou jaciment prehistobric al Pla de Barcelona. *Tribuna d'arqueologia*, 1991, 27–32.

Greenfield, H. J., & Arnold, E. R. (2015). 'Go(a)t milk?' New perspectives on the zooarchaeological evidence for the earliest intensification of dairying in south eastern Europe. *World Archaeology*, 47(5), 792–818. https://doi.org/10.1080/00438243.2015.1029076

Gregg, M. W., Banning, E. B., Gibbs, K., & Slater, G. F. (2009). Subsistence practices and pottery use in Neolithic Jordan: molecular and isotopic evidence. *Journal of Archaeological Science*, 36(4), 937–946. https://doi.org/http://dx.doi.org/10.1016/j.jas.2008.09.009

Gregg, M. W., & Slater, G. F. (2010). A new method for extraction, isolation and transesterification of free fatty acids from archaeological pottery. *Archaeometry*, 52(5), 833–854. https://doi.org/10.1111/j.1475-4754.2010.00518.x

Grivé, M. (1931). L'Esquerda de les Roques de "El Pany" (Penedès). *Anuari de l'Institut d'Estudis Catalans*. (1), 13–33.

Guasch-Jané, M. R., Ibern-Gómez, M., Andrés-Lacueva, C., Jáuregui, O., & Lamuela-Raventós, R. M. (2004). Liquid Chromatography with Mass Spectrometry in Tandem Mode Applied for the Identification of Wine Markers in Residues from Ancient Egyptian Vessels. *Analytical Chemistry*, 76(6), 1672–1677. https://doi.org/10.1021/ac035082z

Guasch-Jané, M. R., Andrés-Lacueva, C., Jáuregui, O., & Lamuela-Raventós, R. M. (2006a). First evidence of white wine in ancient Egypt from Tutankhamun's tomb. *Journal of Archaeological Science*, 33(8), 1075–1080. https://doi.org/http://dx.doi.org/10.1016/j.jas.2005.11.012

Guasch-Jané, M. R., Andrés-Lacueva, C., Jáuregui, O., & Lamuela-Raventós, R. M. (2006b). The origin of the ancient Egyptian drink Shedeh revealed using LC/MS/MS. *Journal of Archaeological Science,* 33(1), 98–101.

Guerra, E., Delibes, G., Rodriguez, J. A., Crespo, M., Gómez, A., Herrán, J. I., ... Matamala, J. (2012). Residuos de productos lácteos y de grasa de carne en dos recipientes cerámicos de la Edad del bronce del valle medio del Duero. *BSAA Arqueología*, LXXVII–LXX, 105–137.

Guerra, E. (2006). Sobre la función y el significado de la cerámica campaniforme a la luz de los análisis de contenidos. *Trabajos De Prehistoria*, 63(1), 69–84.

Guilaine, J. (1976a). La neolitización de las costas mediterráneas de Francia y España. *Cuadernos de Prehistoria y Arqueología Castellonenses*, 3, 39–50.

Guilaine, J. (1976b). *Premiers bergers et paysans de l'Occident méditerranéen. Mouton.* Paris-La-Haye.

Guilaine, J. (1984). Le Néolithique ancien en Languedoc et Catalogne: éléments et réflexions pour un essai de périodisation. In F. Jordá: *oblata scripta praehistorica* (pp. 271–286). Ediciones Universidad de Salamanca.

Guilaine, J., & Marztluff, M. (1995). Les excavacions a la Balma de la Margineda (1979-1991). Momoster d'Afers Socials i Cultura. Govern d'Andorra.

H

Hammann, S., & Cramp, L. J. E. (2018). Towards the detection of dietary cereal processing through absorbed lipid biomarkers in archaeological pottery. *Journal of Archaeological Science*, 93, 74–81. https://doi.org/10.1016/j.jas.2018.02.017

Hammann, S., Cramp, L. J. E., Whittle, M., & Evershed, R. P. (2018). Cholesterol degradation in archaeological pottery mediated by fired clay and fatty acid pro-oxidants. *Tetrahedron Letters*, 59(50), 4401–4404. https://doi.org/10.1016/j.tetlet.2018.10.071

Hansel, F. A., Copley, M. S., Madureira, L. A. S., & Evershed, R. P. (2004). Thermally produced ω-(o-alkylphenyl)alkanoic acids provide evidence for the processing of marine products in archaeological pottery vessels. *Tetrahedron Letters*, 45(14), 2999–3002. https://doi.org/http://dx.doi.org/10.1016/j.tetlet.2004.01.111

Harris, M. (1998). *Good to eat: Riddles of food and culture.* Waveland Press.

Harzbecher, K., & González, J. (2016). El jaciment de Reina Amàlia 31. In A. Gómez & M. Molist (Eds.), *La rehistòria al pla de Barcelona*. (pp. 201–205). Barcelona, Spain: Ajuntament de Barcelona.

Hastorf, C. A., & DeNiro, M. J. (1985). Reconstruction of prehistoric plant production and cooking practices by a new isotopic method. *Nature*, 315(6019), 489–491. https://doi.org/10.1038/315489a0

Hayden, B., Canuel, N., & Shanse, J. (2012). What Was Brewing in the Natufian? An Archaeological Assessment of Brewing Technology in the Epipaleolithic. *Journal of Archaeological Method and Theory*, 20(1), 102–150. https://doi.org/10.1007/s10816-011-9127-y

Heaton, T. H. E. (1999). Spatial, Species, and Temporal Variations in the δ13C/δ12C Ratios of C3 Plants: Implications for Palaeodiet Studies. J*ournal of Archaeological Science*, 26(6), 637–649. https://doi.org/10.1006/jasc.1998.0381

Hendy, J., Colonese, A. C., Franz, I., Fernandes, R., Fischer, R., Orton, D., … Rosenstock, E. (2018). Ancient proteins from ceramic vessels at Çatalhöyük West reveal the hidden cuisine of early farmers. *Nature Communications*, 9(1), 4064. https://doi.org/10.1038/s41467-018-06335-6

Henrickson, E. F., & McDonald, M. M. A. (1983). Ceramic Form and Function: An Ethnographic Search and an Archeological Application. *American Anthropologist*, 85(3), 630–643. https://doi.org/10.1525/aa.1983.85.3.02a00070

Heron, C., & Evershed, R. P. (1993). The Analysis of Organic Residues and the Study of Pottery Use. *Archaeological Method and Theory*, 5, 247–284. https://doi.org/10.2307/20170233

Heron, C., Evershed, R. P., & Goad, L. J. (1991). Effects of migration of soil lipids on organic residues associated with buried potsherds. *Journal of Archaeological Science*, 18(6), 641–659. https://doi.org/http://dx.doi.org/10.1016/0305-4403(91)90027-M

Heron, C., Nemcek, N., Bonfield, K. M., Dixon, D., & Ottaway, B. S. (1994). The chemistry of neolithic beeswax. *Naturwissenschaften*, 81(6), 266–269. https://doi.org/10.1007/BF01131579

Heron, C., Nilsen, G., Stern, B., Craig, O. E., & Nordby, C. (2010). Application of lipid biomarker analysis to evaluate the function of 'slab-lined pits' in Arctic Norway. *Journal of Archaeological Science*, 37(9), 2188–2197. https://doi.org/http://dx.doi.org/10.1016/j.jas.2010.03.016

Heron, Carl, Craig, O. E., Luquin, A., Steele, V. J., Thompson, A., & Piličiauskas, G. (2015). Cooking fish and drinking milk? Patterns in pottery use in the southeastern Baltic, 3300–2400 cal BC. *Journal of Archaeological Science*, 63, 33–43. https://doi.org/10.1016/j.jas.2015.08.002

Heron, C., Shoda, S., Breu, A., Czebreszuk, J., Eley, Y., Gorton, M., ... Craig, O. E. (2016). First molecular and isotopic evidence of millet processing in prehistoric pottery vessels. *Nature: Scientific reports*, (December), 1–9. https://doi.org/10.1038/srep38767

Hjulström, B., Isaksson, S., & Karlsson, C. (2008). Prominent migration period building. Lipid and elemental analyses from an excavation at Alby, Botkyrka Södermanland, Sweden. *Acta Archaeologica*, 79(1), 62–78. https://doi.org/10.1111/j.1600-0390.2008.00109.x

Holden, C., & Mace, R. (1997). Phylogenetic Analysis of the Evolution of Lactose Digestion in Adults. *Human Biology*, 69(5), 605–628. https://doi.org/10.3378/027.081.0609

Horiuchi, A., Miyata, Y., Kamijo, N., Cramp, L., & Evershed, R. P. (2015). A Dietary Study of the Kamegaoka Culture Population during the Final Jomon Period, Japan, Using Stable Isotope and Lipid Analyses of Ceramic Residues. *Radiocarbon*, 57(04), 721–736. https://doi.org/10.2458/azu_rc.57.18455.

Hurst, W. J., Martin, R. A., Tarka, S. M., & Hall, G. D. (1989). Authentication of cocoa in maya vessels using high-performance liquid chromatographic techniques. *Journal of Chromatography A*, 466(C), 279–289. https://doi.org/10.1016/S0021-9673(01)84623-9

Hurst, W. J., Tarka, S. M., Powis, T. G., Valdez, F., & Hester, T. R. (2002). Cacao usage by the earliest Maya civilization. *Nature*, 418(6895), 289–290. https://doi.org/10.1038/418289a

I

Ingram, C. J. E., Mulcare, C. A., Itan, Y., Thomas, M. G., & Swallow, D. M. (2009). Lactose digestion and the evolutionary genetics of lactase persistence. *Human Genetics*, 124(6), 579–591. https://doi.org/10.1007/s00439-008-0593-6

Isaksson, S., & Hallgren, F. (2012). Lipid residue analyses of Early Neolithic funnel-beaker pottery from Skogsmossen, eastern Central Sweden, and the earliest evidence of dairying in Sweden. *Journal of Archaeological Science*, 39(12), 3600–3609. https://doi.org/http://dx.doi.org/10.1016/j.jas.2012.06.018

Isaksson, S., Karlsson, C., & Eriksson, T. (2010). Ergosterol (5, 7, 22-ergostatrien-3??-ol) as a potential biomarker for alcohol fermentation in lipid residues from prehistoric pottery. *Journal of Archaeological Science*, 37(12), 3263–3268. https://doi.org/10.1016/j.jas.2010.07.027.

Itan, Y., Powell, A., Beaumont, M. A., Burger, J., Thomas, M. G., & Tanaka, M. M. (2009). The Origins of Lactase Persistence in Europe. *PLoS Computational Biology*, 5(8). https://doi.org/10.1371/journal.pcbi.1000491

Izzo, F. C., Zendri, E., Bernardi, A., Balliana, E., & Sgobbi, M. (2013). The study of pitch via gas chromatography–mass spectrometry and Fourier-transformed infrared spectroscopy: the case of the Roman amphoras from Monte Poro, Calabria (Italy). *Journal of Archaeological Science*, 40(1), 595–600. https://doi.org/http://dx.doi.org/10.1016/j.jas.2012.06.017

J

Jetter, R., Kunst, L., & A.L., S. (2006). Composition of plant cuticular waxes. In M. Reiderer & C. Müller (Eds.), *Biology of the plant cuticle* (pp. 182–215). Oxford: Blackwell Publishing.

Julia, R., & Riera, S. (2010). Usos del sòl i activitats productives a Barcelona a partir de l'analisi paleoambiental de la llacuna litoral medieval del Pla de Palau. *Quarhis*, (6), 164–177.

Julià, R., & Riera, S. (2012). Proposta d'evolució del front marítim de Barcelona durant l'Holocè, a partir de la integració de dades geotècniques, intervencions arqueològiques i cronologies absolutes. *Quarhis*, (8), 16–32.

K

Keeling, C. D. (1979). The Suess effect: 13Carbon-14Carbon interrelations. *Environment International*, 2(4–6), 229–300. https://doi.org/10.1016/0160-4120(79)90005-9

Kimpe, K., Jacobs, P. A., & Waelkens, M. (2001). Analysis of oil used in late Roman oil lamps with different mass spectrometric techniques revealed the presence of predominantly olive oil together with traces of animal fat. *Journal of Chromatography A*, 937(1–2), 87–95. https://doi.org/10.1016/S0021-9673(01)01304-8

Knights, B. A., Dickson, C. A., Dickson, J. H., & Breeze, D. J. (1983). Evidence concerning the roman military diet at Bearsden, Scotland, in the 2nd Century AD. *Journal of Archaeological Science*, 10(2), 139–152. https://doi.org/10.1016/0305-4403(83)90048-1

Kuksis, A., Child, P., Myher, J. J., Marai, L., Yousef, I. M., & Lewin, P. K. (1978). Bile acids of a 3200-year-old Egyptian mummy. Canadian Journal of *Biochemistry*, 56, 1141–1148.

L

Lacan, M., Keyser, C., Ricaut, F.-X., Brucato, N., Duranthon, F., Guilaine, J., … Ludes, B. (2011). Ancient DNA reveals male diffusion through the Neolithic Mediterranean route. *Proceedings of the National Academy of Sciences*, 108(24), 9788–9791. https://doi.org/10.1073/pnas.1100723108

Lantos, I., Spangenberg, J. E., Giovannetti, M. A., Ratto, N., & Maier, M. S. (2015). Maize consumption in pre-Hispanic south-central Andes: chemical and microscopic evidence from organic residues in archaeological pottery from western Tinogasta (Catamarca, Argentina). *Journal of Archaeological Science*, 55, 83–99. https://doi.org/http://dx.doi.org/10.1016/j.jas.2014.12.022

Laorden, V., de Prada, J., Monseñe, F. J. M., & Serra, J. (1993). L'estudi geològic del solar de la Caserna de Sant Pau. *Tribuna d'arqueologia*, 1991–1992, 33–35.

Leonardi, M. (2014). Lactase persistence and milk consumption in Europe: An interdisciplinary approach involving genetics and archaeology. *Documenta Praehistorica*, 40(1), 85–96. https://doi.org/10.4312/dp.40.8.

Lightfoot, E., Boneva, B., Miracle, P. T., Šlaus, M., & O'connell, T. C. (2011). Exploring the Mesolithic and Neolithic transition in Croatia through isotopic investigations. *Antiquity*, 85(327), 73–86.

Lin, D. S., Conner, W. E., Napton, L. K., & Heizer, R. F. (1978). Steroids of 2000-year-old human coprolite. *Journal of Lipid Research*, 19, 215–221.

Lloveras, L., Vicente, O., Molist, M., Nadal, J., Riera, S., Julià, R., & Estrada, A. (2014). Interpretación tafonómica de la malacofauna marina en el yacimiento neolítico de la Caserna de Sant Pau del Camp (Barcelona). *Archaeofauna*, 23, 169–179.

Lough, A. K. (1977). The phytanic acid content of the lipids of bovine tissues and milk. *Lipids*, 12(1), 115–119.

Lucquin, A., Colonese, A. C., Farrell, T. F. G., & Craig, O. E. (2016a). Utilising phytanic acid diastereomers for the characterisation of archaeological lipid residues in pottery samples. *Tetrahedron Letters*, 57(6), 703–707. https://doi.org/10.1016/j.tetlet.2016.01.011

Lucquin, A., Gibbs, K., Uchiyama, J., Saul, H., Ajimoto, M., Eley, Y., ... Craig, O. E. (2016b). Ancient lipids document continuity in the use of early hunter–gatherer pottery through 9,000 years of Japanese prehistory. *Proceedings of the National Academy of Sciences*, 113(15), 3991–3996. https://doi.org/10.1073/pnas.1522908113

Lucquin, A., Robson, H. K., Eley, Y., Shoda, S., Veltcheva, D., Gibbs, K., ... Craig, O. E. (2018). The impact of environmental change on the use of early pottery by East Asian hunter-gatherers. *Proceedings of the National Academy of Sciences*, 201803782. https://doi.org/10.1073/pnas.1803782115.

M

Malgosa, A. (2016). La població: característiques físiques i estat de salut. In A Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 151–156). Barcelona, Spain: Ajuntament de Barcelona.

Manen, C. (2002). Structure et identité des styles céramiques du Neolithique Ancien entre Rhône et Èbre. *Gallia Préhistoire,* 44, 121–165.

Manning, K, & Conolly, J. (2013a). Animal exploitation in the early Neolithic of the Balkans and central Europe. In S., Colledge, J., Conolly, K., Dobney, K., Manning, S., Shennan. *Origins and Spread of Domestic Animals in Southwest Asia and Europe*, 59, (pp. 237-252). Left Coast Press.

Manning, K., Downey, S. S., Colledge, S., Conolly, J., Stopp, B., Dobney, K., & Shennan, S. (2013b). The origins and spread of stock-keeping: the role of cultural and environmental influences on early Neolithic animal exploitation in Europe. *Antiquity*, 87(338), 1046–1059.

Manzano, E., García, A., Alarcón, E., Cantarero, S., Contreras, F., & Vílchez, J. L. (2015). An integrated multianalytical approach to the reconstruction of daily activities at the Bronze Age settlement in Peñalosa (Jaén, Spain). *Microchemical Journal*, 122, 127–136. https://doi.org/http://dx.doi.org/10.1016/j.microc.2015.04.021

Marcet, R. (1981). El Neolític Antic (Cardial-Epicardial) a Catalunya. In G. Camps, M. Llongueras, R. Marcet, M. A. Petit, & J. Rovira (Eds.), *El Neolític a Catalunya. Taula rodona de Montserrat.* (1a ed., pp. 15–27). Tortosa, Spain: Publicacions de l'Abadia de Montserrat.

Marlasca, R. (2013). El primer Neolítico y la pesca. La Cova de les Cendres como paradigma. In A., Sanchis, J.L., Pascual. *Animals i Arqueologia Hui* (pp. 219–234).

Martí, B., Capel, J., & Juan-Cabanilles, J. (2009). Una forma singular de las cerámicas neolíticas de la Cova de l'Or (Beniarrés, Alicante): los vasos con asa-pitorro. In Colective (Ed.), *De Méditerranée et d'ailleurs... Mélanges offerts à Jean Guilaine* (pp. 463–482). Toulouse: Archives d'Écologie Préhistorique.

Martín, A. (1992). Lectura de la distribució i caracterització dels jaciments cardials. In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 68–71). Puigcerdà, Spain: Institut d'Estudis Ceretans.

Martín, A., & Miret, M. (2011). Més d'un segle de recerques prehistòriques al massís de Garraf. In A. Blasco, M. Edo, & M. J. Villalba (Eds.), *La cova de Can Sadurní i la Prehistòria de Garraf. Recull de 30 anys d'investigació.* (pp. 167–186). Edar. Arqueología y Patrimonio.

Martín, A., Biosca, A., & Albareda, M. J. (1984). Excavacons a la Cova del Frare (Matadepera, Vallès Occidental). Dinàmica ecològica, esqüència cultural i cronologia absoluta. *Tribuna d'arqueologia*, 91–103.

Martín, A., Guilaine, J., & Thommeret, J. Y. (1981). Estratigrafia y dataciones de C14 del yacimiento de la "Cova del Frare" de St. Llorenç del Munt (Matadepera, Barcelona). *Zephyrus*, (XXXII–XXXIII), 101–111.

Martín, A., Martín, J., Villalba, P., & Tresserras, J. (2005). Ca l'Oliaire (Berga, Barcelona), un asentamiento neolítico en el umbral del IV milenio con residuos de sal y de productos lácteos. In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico Peninsular* (pp. 175–185). Santander: Servicio de Publicaciones, Universidad de Cantabria.

Martín, A., Edo, M., Tarrús, J., & Clop, X. (2010). Le Neolithique ancien de Catalogne (VIe - première moitié du Ve millénaire av J.C.) Les séquences chronoculturelles. In C Manen, F. Convertini, D. Binder, & I. Sénépart (Eds.), *Premières sociétés paysannes de Méditerranée occidentale. Structures des productions céramiques.* (pp. 197–214). Toulouse.

Martins, H., Oms Arias, F. X., Pereira, L., Pike, A. W. G., Rowsell, K., & Zilhão, J. (2015). Radiocarbon dating the beginning of the Neolithcis in Iberia: new results, new problems. *Journal of Mediterranean Archaeology*, 28(1), 105–131. https://doi.org/10.1558/jmea.v28i1.27503

Masachs, J. M. (1975). Las armas y útiles de bronce hallados en el Penedés. In *Crónica del XIII Congreso Arqueológico Nacional* (pp. 455–464). Seminario de Arqueología.

Masschelein-Keiner, L. (1989). Les résines de réparation du niveau V. In P. Pétrequin (Ed.), L*es sites littoraux néolithiques de clairvaux-les-lacs (Jura), le Néolithique moyen.* Paris: Maison des Sciences de l'Homme.

Mathe, C., Culioli, G., Archier, P., & Vieillescazes, C. (2004). Characterization of archaeological frankincense by gas chromatography–mass spectrometry. *Journal of Chromatography A*, 1023(2), 277–285. https://doi.org/http://dx.doi.org/10.1016/j.chroma.2003.10.016

Matlova, V., Roffet-Salque, M., Pavlu, I., Kyselka, J., Sedlarova, I., Filip, V., & Evershed, R. P. (2017). Defining pottery use and animal management at the Neolithic site of Bylany (Czech Republic). *Journal of Archaeological Science: Reports*, 14(April), 262–274. https://doi.org/10.1016/j.jasrep.2017.05.028

Maya, J. L., López-Cachero, J., & Tresserras, J. (1999). Nuevos análisis de contenidos en recipientes cerámicos del hábitat de Genó (Aitona, Lleida). In P. Arias, R. Ontañón, & C. García-Moncó (Eds.), *III Congreso del Neolítico en la Peninsula Ibérica* (pp. 27–28). Santander: Monografías del Instituto Internacional de Investigaciones Prehistoricas de Cantabria

Mazar, A., Namdar, D., Panitz-Cohen, N., Neumann, R., & Weiner, S. (2008). Iron Age beehives at Tel Rehov in the Jordan valley. *Antiquity*, 82(317), 629–639. https://doi.org/10.1017/s0003598x00097271

McClure, S. B., Magill, C., Podrug, E., Moore, A. M. T., Harper, T. K., Culleton, B. J., ... Freeman, K. H. (2018). Fatty acid specific δ13C values reveal earliest Mediterranean cheese production 7,200 years ago. *PLoS one*, 13(9), e0202807. https://doi.org/10.1371/journal.pone.0202807

McGovern, P. E., Glusker, D. L., Exner, L. J., & Voigt, M. M. (1996). Neolithic resinated wine. *Nature*, 381(6582), 480–481. https://doi.org/10.1038/381480a0

Mensua, C., & Piqué, R. (2008). L'aprofitament de l'entorn forestal per a l'obtenció de combustible. *Quarhis*, 4(4), 52–53.

Mercadal, O., Aliaga, S., Esteve, X., Mangado, X., Nadal, J., Cubero, C., & Alvarez, R. (2009). L'assentament del neolític antic de les mines de Sanavastre (Sanavastre, Das, La Cerdanya). *Els Pirineus i Les Àrees Circumdants Durant El Tardiglaciar*. Mutacions I.

Mestres, J. (1981). Avançament a l'estudi del jaciment de Guixeres de Vilobí. *Pyrenae*, 17–18, 35–54.

Mestres, J. (1987). La indústria lítica en sílex del neolític antic de les Guixeres de Vilobí. *Olerdulae*, 1-2-3–4, 5–71.

Mestres, J. (1992). Neolitització i territori. In: Estat de la investigació sobre el neolític a Catalunya. In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 72–75). Puigcerdà: Institut d'Estudis Ceretans.

Mestres, J, & Martín, A. (1995). Calibración de las fechas radiocarbónicas y su contribución al estudio del neolítico catalán. In M. Borrell, J. Bosch, & A. Ribes (Eds.), *I Congrés del Neolític a la Península Ibèrica.* (Vol. 1, pp. 791–804). Gavà/Bellaterra: Rubricatum.

Michel, R. H., McGovern, P. E., & Badler, V. R. (1993). The First Wine and Beer. *Analytical Chemistry*, 65(8), 408–413.

Milner, N., Craig, O. E., Bailey, G. N., Pedersen, K., & Andersen, S. H. (2004). Something fishy in the Neolithic? A re-evaluation of stable isotope analysis of Mesolithic and Neolithic coastal populations. *Antiquity*, 78(299), 9–22. https://doi.org/Article

Miró, J. M. (1991). La Fauna del jaciment de Les Guixeres de Vilobí (Alt Penedès) en el context del Neolític Antic a Catalunya. In J. Guilaine, M. Molist, M. A. del Rincon, & J. Vaquer (Eds.), *9è Col·loqui Internacional d'Arqueologia de Puigcerdà. Estat de la investigació sobre el neolític a Catalunya* (pp. 79–84). Puigcerdà: Institut d'Estudis Ceretans.

Mitkidou, S., Dimitrakoudi, E., Urem-Kotsou, D., Papadopoulou, D., Kotsakis, K., Stratis, J. A., & Stephanidou-Stephanatou, I. (2008). Organic residue analysis of Neolithic pottery from North Greece. *Microchimica Acta*, 160(4), 493–498. https://doi.org/10.1007/s00604-007-0811-2

Molina, E. (2011). El artefacto en vida. Integración del análisis de residuos orgánicos en el estudio de la producción alfarera. *Estrat Crític*, 2(5), 244–257.

Molina, E. (2015a). Análisis de residuos orgánicos de los vasos del Hipogeu de la Sagrera. In M. Molist (Ed.), *Projecte d'estudi de les restes de l'hipogeu de la Sagrera. Fase II.* Projecte Prehistòria de Barcelona. UAB-Servei d'Arqueologia. Ajuntament de Barcelona. Unpublished.

Molina, E. (2015b). *La producción cerámica en el sudeste de la península ibérica Durante el III y II milenio ane (2200-1550 cal ane): integración del análisis de residuos orgánicos en la caracterización funcional de los recipientes argáricos.* PhD Thesis Universitat Autònoma de Barcelona, Barcelona.

Molina, L., Bernabeu, J., & García, P. (2010). Méthode d'analyse stylistique des céramiques du Néolithique ancien cardial en Pays valencien (Espagne). In C Manen, F. Convertini, D. Binder, & I. Sénépart (Eds.), *Premières sociétés paysannes de Méditerranée occidentale. Structures des productions céramiques.* (1st ed., Vol. Memoire LI, pp. 65–77). Toulouse, France: Société Prehistorique Française.

Molinas, R. (2009). *Estudi Arqueozoològic sobre la gestió dels recursos animals al jaciment del Carrer Reina Amàlia (Barcelona)*. Barcelona. Unpublished report.

Molist, M. (2006). El jaciment prehistòric de Sant Pau del Camp. *Quarhis*, (2), 180–181.

Molist, M. (2009). Prehistòria del pla de Barcelona. *Quarhis*, (5), 227–230.

Molist, M., & Gómez, A. (2016). El Jaciment de la Caserna de Sant Pau del Camp. In A. Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 173–178). Barcelona, Spain: Ajuntament de Barcelona.

Molist, M., Vicente, O., & Farré, R. (2008). El jaciment de la caserna de Sant Pau del Camp: aproximació a la caracterització d'un assentament del neolític antic. *Quarhis*, (4), 14–24.

Molist, M., Bosch, J., Clop, X., Gómez, A., & Guerrero, E. (2009). Des vases cardiales dans la ville de Barcelone: Apports du site de la Caserna de Sant Pau del Camp sur le néolithique ancien de la Catalone. In *e Méditerranée et d'ailleurs...: mélanges offerts à Jean Guilaine.* (pp. 547–555). Tolouse.

Molist, M., Gómez, A., Breu, A., González, J., Bordas, A., Harzbecher, K., & Fernández, J. (2016). Buried structures in Barcelona plain's neolithic settlements: A general revision, morphological and morphometric analysis gaining a functional interpretation. *Quaternary International*, 1–13. https://doi.org/10.1016/j.quaint.2017.05.048

Mora, R., Benito-Calvo, A., Martínez-Moreno, J., Marcen, P. G., & de la Torre, I. (2011). Chrono-stratigraphy of the Upper Pleistocene and Holocene archaeological sequence in Cova Gran (south-eastern Pre-Pyrenees, Iberian Peninsula). *Journal of Quaternary Science*, 26(6), 635–644. https://doi.org/10.1002/jqs.1486

Morales, J., Cebrià, A., Mestres, J., Oms, F., & Allué, E. (2013). La cova de la Guineu. 12000 anys de presència humana a les capçaleres del Foix. In J. Hernández, J. Grau, & J. Melero (Eds.), *III Monografies del Foix* (pp. 172–183). Barcelona: Diputació de Barcelona.

Morgan, E. D., Titus, L., Small, R. J., & Edwards, C. (1984). Gas Chromatographic Analysis Of Fatty Material From A Thule Midden. *Archaeometry*, 26(1), 43–48. https://doi.org/10.1111/j.1475-4754.1984.tb00316.x

Mottram, H. R., Dudd, S. N., Lawrence, G. J., Stott, A. W., & Evershed, R. P. (1999). New chromatographic, mass spectrometric and stable isotope approaches to the classification of degraded animal fats preserved in archaeological pottery. *Journal of Chromatography A*, 833(2), 209–221. https://doi.org/http://dx.doi.org/10.1016/S0021-9673(98)01041-3

Mukherjee, A. J., Copley, M. S., Berstan, R., Clark, K. A., & Evershed, R. P. (2005). Interpretation of δ^{13}C values of fatty acids in relation to animal husbandry, food processing and consumption in prehistory. In J. Mulville & A. K. Outram (Eds.), 9th ICAZ Conference, Durham 2002. *The Zooarchaeology of Fats, Oils, Milk and Dairying.* (pp. 77–93). United Kingdom: Oxbow Book.

Mukherjee, A. J., Gibson, A. M., & Evershed, R. P. (2008). Trends in pig product processing at British Neolithic Grooved Ware sites traced through organic residues in potsherds. *Journal of Archaeological Science*, 35(7), 2059–2073. https://doi.org/http://dx.doi.org/10.1016/j.jas.2008.01.010

Mukherjee, A. J., Berstan, R., Copley, M. S., Gibson, A. M., & Evershed, R. P. (2007). Compound-specific stable carbon isotopic detection of pig product processing in British Late Neolithic pottery. *Antiquity*, 81(313), 743–754. https://doi.org/10.1017/S0003598X00095703

Muñoz, V., & Martínez, P. (2014). L'estudi dels materials ceràmics prehistòrics del Pla del Serrador (Les Franqueses del Vallès, Vallès oriental). *Cypsela*, (19), 159–178.

Mylona, D. (2014). Aquatic animal resources in Prehistoric Aegean, Greece. *Journal of Biological Research (Greece)*, 21(1), 1–11. https://doi.org/10.1186/2241-5793-21-2

N

Nadal, J. (2016). Els recursos marins. In Anna Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 105–109). Barcelona, Spain: Ajuntament de Barcelona.

Nadal, J., & Estrada, A. (2008). Estudi de les restes malacofaunístiques del jaciment de la Caserna de Sant Pau. *Quarhis*, 4, 64–69.

Nadal J., Senabre, M. R., Mestres, J., & Cebria, A. (1999). Evolucion Del Aprovechamiento De Los Recursos Fanisticos Durante El Neolitico En La Comarca De L'Alt Penedes (Barcelona). *SAGVNTUM*, 2(2 EXTRA), 85–93.

Nadal, J., Brieva, E., Gómez, A., Molist, M., González, J., Harzbecher, K., & Lloveras, L. (2015). Nuevos datos para la interpretación de los conjuntos malacofaunísticos de los asentamientos neolíticos en la ciudad de Barcelona: el yacimiento de Reina Amàlia no 31-33. In I. Gutierrez, D. Cuenca, & M. R. Gonzáelz (Eds.), *La investigación Arqueomalacológica en la Península Ibérica: Nuevas Aportaciones.* IV Reunión Científica Arqueomalacología de la Península Ibérica. (pp. 113–124).

Namdar, D., Neumann, R., Goren, Y., & Weiner, S. (2009). The contents of unusual cone-shaped vessels (cornets) from the Chalcolithic of the southern Levant. *Journal of Archaeological Science*, 36(3), 629–636. https://doi.org/http://dx.doi.org/10.1016/j.jas.2008.10.004

Namdar, D., Amrani, A., Getzov, N., & Milevski, I. (2015). Olive oil storage during the fifth and sixth millennia BC at Ein Zippori, Northern Israel. Israel *Journal of Plant Sciences*, 62(1–2), 65–74. https://doi.org/10.1080/07929978.2014.960733

Navarrete, V. (2017). *Domesticación animal y primeras prácticas ganaderas en el noreste peninsular (5500-4500 cal BC). Integración de los análisis bioquímicos en arqueozoología.* PhD Thesis. Departament de Prehistòria. Universitat Autònoma de Barcelona.

Navarrete, V., & Saña, M. (2015). *Informe del análisis arqueozoológico del yacimiento de "Reina Amàlia" (Barcelona) Campañas de excavación arqueológica 2003-2009.* Unpublished report. Barcelona.

Navarrete, V., Colonese, A. C., Tornero, C., Antolín, F., Von Tersch, M., Eulàlia Subirà, M., ... Saña, M. (2017). Feeding Management Strategies among the Early Neolithic Pigs in the NE of the Iberian Peninsula. *International Journal of Osteoarchaeology*, 27(5), 839–852. https://doi.org/10.1002/oa.2598

Needham, S., & Evans, J. (1987). Honey And Dripping: Neolithic Food Residues. *Oxford Journal of Archaeology*, 6(1), 21–28.

Newsome, S. D., Phillips, D. L., Culleton, B. J., Guilderson, T. P., & Koch, P. L. (2004). Dietary reconstruction of an early to middle Holocene human population from the central California coast: Insights from advanced stable isotope mixing models. *Journal of Archaeological Science*, 31(8), 1101–1115. https://doi.org/10.1016/j.jas.2004.02.001

O

Oehlert, A. M., & Swart, P. K. (2014). Interpreting carbonate and organic carbon isotope covariance in the sedimentary record. *Nature Communications*, 5(1), 4672. https://doi.org/10.1038/ncomms5672

Olalde, I., Brace, S., Allentoft, M. E., Armit, I., Kristiansen, K., Booth, T., ... Reich, D. (2018). The Beaker phenomenon and the genomic transformation of northwest Europe. *Nature*, 555(7695), 190–196. https://doi.org/10.1038/nature25738

Olalde, I., Mallick, S., Patterson, N., Rohland, N., Villalba-Mouco, V., Silva, M., ... Reich, D. (2019). The genomic history of the Iberian Peninsula over the past 8000 years. *Science*, 363(6432), 1230–1234. https://doi.org/10.1126/science.aav4040

Oliva, M. (2015). *Aprofitament i transformació de matèries primeres per a l'elaboració d'ornaments durant la prehistòria recent (5600-3400 cal. ane) al nord-est de la península Ibèrica.* PhD Thesis. Departament de Prehistòria. Universitat Autònoma de Barcelona.

Oliva, M., Palomo, A., Terrats, N., Carlús Martín, X., López Cachero, J., & Rodriguez, A. (2008). Las estructuras neolíticas de Can Roqueta (Sabadell, Barcelona). In M. S. Hernández, J. A. Soler, & J. A. López (Eds.), *IV Congreso del Neolítico Peninsular* (Vol. 1, pp. 157–167). Alicante: Museo Arqueológico de Alicante.

Oltra, J. (2010). *La Producció d'aliments al Sud Est de la Península Ibèrica (c. 1500-900 cal ANE)*. PhD thesis. Department of Prehistory. Universitat Autònoma de Barcelona.

Oms, F. X. (2008). Caracterització tècnica, tipològica i cronològica de les ceràmiques del Neolític Antic de la Cova Colomera (Prepirineu de Lleida). *Archivo de Prehistoria Levantina*, XXVII, 51–80.

Oms, F. X. (2014). *La neolitització del Nord-Est de la Península Ibèrica a partir de les datacions de ^{14}C i les primeres ceràmiques impreses. C. 5600-4900 Cal. BC.* PhD Thesis. Universitat de Barcelona.

Oms, F. X. (2017a). *La neolitització del nord-est de la península Ibèrica. Barcelon: Societat Catalana d'Arqueologia*. Premi d'Arqueologia Memorial Josep Barberà i Farràs. Catorzena Edició.

Oms, F. X. (2017b). Fases y territorios de la neolitización del NE de la Península Ibérica ca. 5600-4900 cal BC. *Munibe Antropologia-Arkeologia*, 68(January). https://doi.org/10.21630/maa.2017.68.01

Oms, F. X., & Morales, J. I. (2008). La indústria lítica. In Francesc Xavier Oms & J. I. Morales (Eds.), *Cambrils al Neolític* (pp. 115–195). Cambrils, Spain: Ajuntament de Cambrils.

Oms, F. X., Bargalló Ferrerons, A., Chaler, M., Fontanals Torroja, M., García García, M. S., Manuel López, J., ... Vergès Bosch, J. M. (2008). La Cova Colomera (Sant Esteve de la Sarga, Lleida), una cueva-redil en el prepirineo de Lérida. Primeros resultados y perspectivas de futuro. In M. S. Hernández, J. A. Soler, & J. A. López (Eds.), *IV Congreso del Neolítico Peninsular* (Vol. 1, pp. 230–236). Alicante: Museo Arqueológico de Alicante.

Oms, F. X., Cebrià, A., Morales, J. I., & Pedro, M. (2011). Una inhumació cardial a la cova Foradada (Calafell, Baix Penedès). *Jornades d'Arqueologia Del Penedès, 117–124.*

Oms, F. X., Esteve, X., Mestres, J., Martín, P., & Martins, H. (2014). La neolitización del nordeste de la Península Ibérica: datos radiocarbónicos y culturales de los asentamientos al aire libre del Penedès. *Trabajos de Prehistoria*, 71(1), 42–55. https://doi.org/10.3989/tp.2014.12123

Oms, F. X., Mestres, J., Cebrià, A., Morales, J., Nadal, J., Pedro, M., ... Fullola, J. (2016a). La cova de la Guineu (Font-Rubí , Barcelona) i les relacions plana-muntanya al Penedès durant el neolític inicial. In *Del neolític a l'edat del bronze en el Mediterrani occidental. Estudis en homenatge a Bernat Martí Oliver.* (Vol. TV SIP 119, pp. 97–107). Valencia.

Oms, F. X., Petit, M. Á., & López-Cachero, F. J. (2016b). Motius figuratius en la ceràmica del neolític antic inicial i art (macro)esquemàtic a Catalunya. In *Del Neolític a l'edat Del Bronze En El Mediterrani Occidental. Estudis En Homenatge a Bernat Martí Oliver*, (Vol. TV SIP 491–499). Valencia.

Oms, F. X., Martín, A., Esteve, X., Mestres, J., Morell, B., Subirà, M. E., & Gibaja, J. F. (2016c). The neolithic in northeast iberia: Chronocultural phases and 14C. *Radiocarbon*, 58(2), 291–309. https://doi.org/10.1017/RDC.2015.14

Oms, F. X., Daura, J., Sanz, M., Mendiela, S., Pedro, M., & Martínez, P. (2017). First Evidence of Collective Human Inhumation from the Cardial Neolithic (Cova Bonica, Barcelona, NE Iberian Peninsula). *Journal of Field Archaeology*, 42(1), 43–53. https://doi.org/10.1080/00934690.2016.1260407

Oms, F. X., Mestres, J., Cebrià, A., Morales, J. I., Mendiela, S., Esteve, X., … Fullola, J. M. (in press). Movilidad y fases de ocupación del territorio durante el Neolítico inicial en el Penedès (Barcelona). In: *VI Congreso del Neolítico en la Peninsula Ibérica.* Universidad de Granada.

Oras, E., Lucquin, A., Lõugas, L., Tõrv, M., Kriiska, A., & Craig, O. E. (2017). The adoption of pottery by north-east European hunter-gatherers: Evidence from lipid residue analysis. *Journal of Archaeological Science*, 78, 112–119. https://doi.org/10.1016/j.jas.2016.11.010

Ortman, S. G., Varien, M. D., & Gripp, T. L. (2007). Empirical Bayesian methods for archaeological survey data: an application from the Mesa Verde region. *American Antiquity*, 72(2), 241–272.

Oudemans, T. F. M., & Boon, J. J. (1991). Molecular archaeology: Analysis of charred (food) remains from prehistoric pottery by pyrolysis—gas chromatography/mass spectrometry. *Journal of Analytical and Applied Pyrolysis*, 20, 197–227. https://doi.org/10.1016/0165-2370(91)80073-H

Outram, A. K., Stear, N. A., Kasparov, A., Usmanova, E., Varfolomeev, V., & Evershed, R. P. (2011). Horses for the dead: funerary foodways in Bronze Age Kazakhstan. *Antiquity*, 85(327), 116–128. https://doi.org/doi:10.1017/S0003598X00067478

P

Palomo, A. (2012). *Tecnologia lítica i de la fusta de la prehistòria recent al nord-est peninsular. Anàlisi tecnomorfològica i experimental*. PhD Thesis.Universitat Autònoma de Barcelona.

Palomo, A., Gibaja, J. F., Pique, R., Bosch, A., Chinchilla, J., & Tarru, J. (2011). Harvesting cereals and other plants in Neolithic Iberia: The assemblage from the lake settlement at la Draga. *Antiquity*, 85(329), 759–771. https://doi.org/10.1017/S0003598X00068290

Palomo, A., Piqué, R., Terradas, X., Bosch, À., Buxó, R., Chinchilla, J., … Tarrús, J. (2014). Prehistoric Occupation of Banyoles Lakeshore: Results of Recent Excavations at La Draga Site, Girona, Spain. *Journal of Wetland Archaeology*, 14(1), 58–73. https://doi.org/10.1179/1473297114Z.00000000010.

Palomo, A., Terradas, X., Piqué, R., Rosill, R., Bogdanovic, I., Bosch, À., ... Revelles, J. (2018). Les Coves del Fem (Ulldemolins, Catalunya). *Tribuna d'arqueologia*, 88–103.

Papakosta, V., Oras, E., & Isaksson, S. (2019). Early pottery use across the Baltic–A comparative lipid residue study on Ertebølle and Narva ceramics from coastal hunter-gatherer sites in southern Scandinavia, northern Germany and Estonia. *Journal of Archaeological Science: Reports*, 24, 142–151.

Papakosta, V., Smittenberg, R. H., Gibbs, K., Jordan, P., & Isaksson, S. (2015). Extraction and derivatization of absorbed lipid residues from very small and very old samples of ceramic potsherds for molecular analysis by gas chromatography-mass spectrometry (GC-MS) and single compound stable carbon isotope analysis by gas chromatogra. *Microchemical Journal*, 123, 196–200. https://doi.org/10.1016/j.microc.2015.06.013

Parnell, A. C., Inger, R., Bearhop, S., & Jackson, A. L. (2010). Source partitioning using stable isotopes: Coping with too much variation. *PLoS ONE*, 5(3), 1–5. https://doi.org/10.1371/journal.pone.0009672

Parnell, A. C., Phillips, D. L., Bearhop, S., Semmens, B. X., Ward, E. J., Moore, J. W., ... Inger, R. (2013). Bayesian stable isotope mixing models. *Environmetrics*, 24(6), 387–399. https://doi.org/10.1002/env.2221

Parras, D., Sánchez, A., Ramos, N., Rodríguez, M. O., & Tuñón, J. A. (2011). Identification of fats and beeswax in ceramic vessels of tomb 121 of Castellón Alto (Galera, Granada). *Coalition*, (22), 7–13.

Parras, D., Sánchez, A., Tuñón, J. A., Montejo, M., Ramos, N., & Cañabate, M. L. (2015). Análisis físico-químicos en el santuario de El Pajarillo (Huelma, Jaén) y en el oppidum de Puente Tablas (Jaén). In A. Ruiz & M. Molinero (Eds.), *Jaén, tierra ibera: 40 años de investigación y transferencia* (1a ed., pp. 137–143). Jaén: Universidad de Jaén. Servicio de Publicaciones.

Paul, D., Skrzypek, G., & Fórizs, I. (2007). Normalization of measured stable isotopic compositions to isotope reference scales - a review. *Rapid Communications in Mass Spectrometry*, 21, 30006–33014.

Pecci, A., Clarke, J., Thomas, M., Muslin, J., van der Graaff, I., Toniolo, L., … Di Pasquale, G. (2017). Use and reuse of amphorae. Wine residues in Dressel 2–4 amphorae from Oplontis Villa B (Torre Annunziata, Italy). *Journal of Archaeological Science: Reports*, 12. https://doi.org/10.1016/j.jasrep.2017.02.025

Pereira, I. (2019). Carrer de Berenguer de Palou, 52-62. *Anuari d'Arqueologia i Patrimoni de Barcelona*, 2017, 36–38.

Petit, M. A., Estrada, A., Bergadà, M. M., Fullola, J. M., Bartrolí, R., Gamarra, A., … Farell, D. (1996). El procés de neolitització a la vall del Segre. La cova del Parco (Alòs de Balaguer, la Noguera). *Estudi de les ocupacions humanes del Vè al IIn mil·lenni aC*. SERP/Universitat de Barcelona, 70.

Phillips, D. L. (2012). Converting isotope values to diet composition: the use of mixing models. *Journal of Mammalogy*, 93(2), 342–352. https://doi.org/10.1644/11-MAMM-S-158.1

Phillips, D. L., & Gregg, J. W. (2001). Uncertainty in source partitioning using stable isotopes. *Oecologia*, 127(2), 171–179. https://doi.org/10.1007/s004420000578

Phillips, D. L., & Gregg, J. W. (2003). Source partitioning using stable isotopes: Coping with too many sources. *Oecologia*, 136(2), 261–269. https://doi.org/10.1007/s00442-003-1218-3

Phillips, D. L., Inger, R., Bearhop, S., Jackson, A. L., Moore, J. W., Parnell, A. C., … Ward, E. J. (2014). Best practices for use of stable isotope mixing models in food-web studies. *Canadian Journal of Zoology*, 92(10), 823–835. https://doi.org/10.1139/cjz-2014-0127

Phillips, D. L., Newsome, S. D., & Gregg, J. W. (2005). Combining sources in stable isotope mixing models: Alternative methods. *Oecologia*, 144(4), 520–527. https://doi.org/10.1007/s00442-004-1816-8

Piera, M. (2010a). *Memòria de la prospecció arqueològica a l'entorn del Polígon "El Molló" (Mora la Nova i Tivissa).* Unpublished report.

Piera, M. (2010b). El jaciment neolític del Pla del Gardelo (Juneda). Cabal de Petjades. In *Cabal de petjades VII Trobada d'Estudiosos de Les Garrigues*, Vinaixa, 24/10/2009, 59–67. Editorial Fonoll.

Piera, M., Gómez, A., Molist, M., Ríos, P., Alàntara, R., Arnaiz, R., & Montero, I. (2016). El tram baix de l'Ebre a les èpoques del Neolític i Bronze inicil: Aportacions al seu coneixement a partir de l'assentament del Molló (Móra la Nova). In F. Balde, N. Pegueroles, J. Martínez, J. Diloli, M. del M. Villalbí, R. Ferré, ... M. Poy (Eds.), *I Jornades d'Arqueologia de les terres de l'Ebre* (pp. 91–104). Tortosa, España: Serveis Territorials de Cultura de les Terres de l'Ebre.

Piqué, R., Revelles, J., Burjachs, F., Caruso Fermé, L., & Pérez-Obiol, R. (2018). Interdisciplinary approach to the landscape and firewood exploitation during the Holocene at La Garrotxa (Girona, NE Iberia). *Quaternary International*, 463, 401–413. https://doi.org/10.1016/j.quaint.2016.11.025

Piqué, R., Palomo, A., Terradas, X., Tarrús, J., Buxó, R., Bosch, À., ... Saña, M. (2015). Characterizing prehistoric archery: technical and functional analyses of the Neolithic bows from La Draga (NE Iberian Peninsula). *Journal of Archaeological Science*, 55(0), 166–173. https://doi.org/http://dx.doi.org/10.1016/j.jas.2015.01.005

Plantinga, T. S., Alonso, S., Izagirre, N., Hervella, M., Fregel, R., van der Meer, J. W. M., ... de la Rúa, C. (2012). Low prevalence of lactase persistence in Neolithic South-West Europe. *European Journal of Human Genetics : EJHG*, 20(7), 778–782. https://doi.org/10.1038/ejhg.2011.254

Pollard, A. M., & Heron, C. (2008). The Chemistry and Use of Resinous Substances. In A.M. Pollard, C. Heron. *Archaeological Chemistry*. (pp. 235-269) The Royal Society of Chemistry. Cambridge.

Pou, R., & Martí, M. (2005). Les restes del neolític antic al carrer d'en Xammar (Mataró, Maresme). *Laietania*, (16), 9–24.

Pou, R., Martí, M., Jordana, X., Malgosa, A., & Gibaja, J. F. (2010). L'enterrament del neolític antic de la Plaça de la Vila de Madrid (Barcelona). Una estructura funeraria del VIè mil. leni aC. *Quarhis*, 6, 94–107.

Powell, J. (2003). The fish bone assemblage from the Cave of Cyclope, Youra: evidence for continuity and change. *British School at Athens Studies*, 173–179.

Prieto, M. P., Tresserras, J., & Matamala, J. (2005). Ceramic production in the northwestern Iberian Peninsula: studying the functional features of pottery by analyzing organic material. *Trabalhos de Arqueología*, 42, 193–200.

Prieto, M. P., Tresserras, J., & Matamala, J. (2009). ¿Qué se comía en la antiguedad en Galícia? In M. P. Prieto & F. Criado-Boado (Eds.), *Reconstruyendo la historia de la comarca del Ulla-Deza* (Galicia, España) (1a ed., pp. 129–134). Santiago de Compostela: Consejo Superior de Investigaciones Científicas. Laboratorio de Arqueoloxía do Instituto de Estudos Galegos Padre Armiento. Xunta de Galicia.

R

Radi, G., & Wilkens, B. (1989). Il sito a Ceramica Impressa di Santo Stefano (Ortucchio, l'Aquila), Notizia Preliminare. In I. Giglio (Ed.), *Rassegna di Archeologia* (pp. 97–117). Firenze, Italy: Associazione Archaeologica Piombinese.

Rageot, M., Pêche-Quilichini, K., Py, V., Filippi, J. J., Fernandez, X., & Regert, M. (2015). Exploitation of Beehive Products, Plant Exudates and Tars in Corsica During the Early Iron Age. *Archaeometry*, (Published online: 19 May 2015), 58(2), 315-332. https://doi.org/10.1111/arcm.12172

Rainsford, C., O'Connor, T., & Miracle, P. (2014). Fishing in the Adriatic at the Mesolithic–Neolithic transition: Evidence from Vela Spila, Croatia. *Environmental Archaeology*, 19(3), 311–320.

Ramsey, C. B. (2009). Bayesian Analysis of Radiocarbon Dates. *Radiocarbon*, 51(1), 337–360.

Raven, A. M., van Bergen, P. F., Stott, A. W., Dudd, S. N., & Evershed, R. P. (1997). Formation of long-chain ketones in archaeological pottery vessels by pyrolysis of acyl lipids. *Journal of Analytical and Applied Pyrolysis*, 40–41(0), 267–285. https://doi.org/http://dx.doi.org/10.1016/S0165-2370(97)00036-3

Reber, E. A., & Evershed, R. P. (2004a). How Did Mississippians Prepare Maize? The Application of Compound-Specific Carbon Isotope Analysis to Absorbed Pottery Residues From Several Mississippi Valley Sites. *Archaeometry*, 46(1), 19–33. https://doi.org/10.1111/j.1475-4754.2004.00141.x

Reber, E. A., & Evershed, R. P. (2004b). Identification of maize in absorbed organic residues: a cautionary tale. J*ournal of Archaeological Science*, 31(4), 399–410. https://doi.org/http://dx.doi.org/10.1016/j.jas.2003.09.008

Reber, E. A., & Hart, J. P. (2008). Pine resins and pottery sealing: Analysis of absorbed and visible pottery residues from central new york state. *Archaeometry,* 50(6), 999–1017. https://doi.org/10.1111/j.1475-4754.2008.00387.x

Reber, E. A., Dudd, S. N., van der Merwe, N. J., & Evershed, R. P. (2004). Direct detection of maize in pottery residues via compound specific stable carbon isotope analysis. *Antiquity*, 78(301), 682–691. https://doi.org/doi:10.1017/S0003598X00113316

Reber, E. A., Baumann, T. E., Monaghan, G. W., & Myers, K. N. (2015). Absorbed Residue Analysis of a Mississippi Plain Jar from Angel Mounds (12Vg1). *Advances in Archaeological Practice*, 3(01), 29–49. https://doi.org/10.7183/2326-3768.3.1.29

Reber, E. A., Kerr, M. T., Whelton, H. L., & Evershed, R. P. (2018). Lipid Residues from Low-Fired Pottery. *Archaeometry*, 61(1), 131–144. https://doi.org/10.1111/arcm.12403

Regert, M. (2004). Investigating the history of prehistoric glues by gas chromatography-mass spectrometry. *Journal of Separation Science*, 27(3), 244–254. https://doi.org/10.1002/jssc.200301608

Regert, M. (2011). Analytical strategies for discriminating archaeological fatty substances from animal origin. *Mass Spectrometry Reviews*, 30, 177–220.

Regert, M., Bland, H. A., Dudd, S. N., Bergen, P. F. van, & Evershed, R. P. (1998). Free and bound fatty acid oxidation products in archaeological ceramic vessels. *Proceedings of the Royal Society B: Biological Sciences*, 265(1409), 2027–2032. https://doi.org/10.1098/rspb.1998.0536

Regert, M., Garnier, N., Binder, D., & Pétrequin, P. (2000). Les adhésifs néolithiques : quels matériaux utilisés, quelles techniques de production dans quel contexte social ? L'exemple des adhésifs des sites de Giribaldi et de Chalain. In *XXèmes Rencontres Internationales d'Archéologie et d'Histoire d'Antibes*. (pp.586-604) Antibes, France.

Regert, M., Colinart, S., Degrand, L., & Decavallas, O. (2001). Chemical alteration and use of beeswax through time: accelerated ageing tests and analysis of archaeological samples from various environmental contexts. *Archaeometry,* 43(4), 549–569. https://doi.org/10.1111/1475-4754.00036

Regert, M., Devièse, T., Le Hô, A. S., & Rougeulle, A. (2008). Reconstructing ancient yemeni commercial routes during the middle ages using structural characterization of terpenoid resins. *Archaeometry*, 50(4), 668–695. https://doi.org/10.1111/j.1475-4754.2007.00372.x

Revelles, J., Burjachs, F., Palomo, A., Piqué, R., Iriarte, E., Pérez-Obiol, R., & Terradas, X. (2018). Human-environment interaction during the Mesolithic-Neolithic transition in the NE Iberian Peninsula. Vegetation history, climate change and human impact during the Early-Middle Holocene in the Eastern Pre-Pyrenees. *Quaternary Science Reviews*, 184(September), 183–200. https://doi.org/10.1016/j.quascirev.2017.08.025

Revelles, J. (2017). Archaeoecology of Neolithisation. Human-environment interactions in the NE Iberian Peninsula during the Early Neolithic. J*ournal of Archaeological Science: Reports*, 15, 437–445. https://doi.org/10.1016/j.jasrep.2016.02.004

Richards, M. P., & Schulting, R. (2006). Touch not the fish : the Mesolithic-Against the grain ? A response. *Antiquity*, 80(308), 444–456.

Rieley, G. (1994). Derivatization of organic compounds prior to gas chromatographic-combustion-isotope ratio mass spectrometric analysis: identification of isotope fractionation processes. *Analyst*, 119(5), 915–919. https://doi.org/10.1039/AN9941900915

Riera, S., Julià, R., & Picornell, L. (2016). Evolució del paleoambient de la plana barcelonina al Neolític. In A. Gomez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 65–73). Barcelona, Spain: Ajuntament de Barcelona.

Riera, S. (1994). Evolució del paisatge vegetal holocè al Pla de Barcelona, a partir de les dades pol·líniques. PhD Thesis. Departament de Prehistòria, Història Antiga i Arqueologia. Universitat de Barcelona, Barcelona.

Rios, P., García, R., Aliaga, R., & Blanco, J. F. (2011). Las cerámicas: caracterización y contenido. In C. Blasco, C. Liesau, & P. Ríos (Eds.), *Yacimientos Calcolíticos con campaniforme de la región de Madrid: nuevos estudios* (pp. 319–346). Madrid: Universidad Autónoma de Madrid.

Rojo-Guerra, M., Garrido-Pena, R., & García-Martínez, Í. (2008). No solo cerveza. Nuevos tipos de bebidas alcohólicas identificados en análisis de contenidos de cerámicas campaniformes del Valle de Ambrona (Soria). *CPAG*, (18), 91–105.

Romanus, K., Van Neer, W., Marinova, E., Verbeke, K., Luypaerts, A., Accardo, S., ... Waelkens, M. (2008). Brassicaceae seed oil identified as illuminant in Nilotic shells from a first millennium AD Coptic church in Bawit, Egypt. *Analytical and Bioanalytical Chemistry*, 390(2), 783–793. https://doi.org/10.1007/s00216-007-1704-2

Rosada, L., Manhita, A., Tavares, D., Alfenim, R., Mirâa, J., & Barrocas-Dias, C. (2014). Análise de resíduos orgânicos em cerâmicas do Depósito Votivo de Garvâo, 2o Idade do Ferro. In D. Juanes & C. Roldán (Eds.), *X Congreso Ibérico de arqueometría*. Valencia: Subdirección de Conservación, Restauración e Investigación IVC+R de CulturArts Generalitat.

Rottländer, R. C. A., & Schlichtherle, H. (1983). Chemical analysis of fat residues in prehistoric vessels. *Naturwissenschaften*, 70(1), 33–38.

Rovira I Port, J. (1976). El Neolitic a la Cerdanya i Alta Vall del Segre in 1r Col. loqui Internacional d'Arqueologia de Puigcerdà. *Cypsela*, 1, 39–48.

S

Salazar-García, D. C. (2009). Estudio de la dieta en la población neolítica de Costamar. Resultados preliminares de análisis de isótopos estables de carbono y nitrógeno. In *Torre La Sal (Ribera de Cabanes, Castellón). Evolución del paisaje antrópico desde la prehistoria hasta el medioevo.* (pp. 411–418).

Salazar-García, D. C. (2014a). Estudi de la dieta en la població de Cova dels Diablets mitjançant anàlisi d'isòtops estables del carboni i del nitrògen en col·làgen ossi. Resultats preliminars. In G. Aguilella, D. Roman, & P. García Borja (Eds.), *La cova dels diablets (Alcalá de Xivert, Castelló). Prehistoria a la Serra d'Irta* (p. 102). Servei d'Investigacions Arqueològiques i Prehistòriques, Diputació de Castelló.

Salazar-García, D. C., Aura, J. E., Olària, C. R., Talamo, S., Morales, J. V., & Richards, M. P. (2014b). Isotope evidence for the use of marine resources in the Eastern iberian mesolithic. *Journal of Archaeological Science*, 42(1), 231–240. https://doi.org/10.1016/j.jas.2013.11.006

Salazar-García, D. C., Fontanals-Coll, M., Goude, G., & Subirà, M. E. (2018). "To 'seafood' or not to 'seafood'?" An isotopic perspective on dietary preferences at the Mesolithic-Neolithic transition in the Western Mediterranean. *Quaternary International*, 470, 497–510. https://doi.org/10.1016/j.quaint.2017.12.039

Salque, M., Radi, G., Tagliacozzo, A., Uria, B. P., Wolfram, S., Hohle, I., … Evershed, R. P. (2012). New insights into the Early Neolithic economy and management of animals in Southern and Central Europe revealed using lipid residue analyses of pottery vessels. *Anthropozoologica*, 47(2), 45–62. https://doi.org/10.5252/az2012n2a4

Salque, M., Bogucki, P. I., Pyzel, J., Sobkowiak-Tabaka, I., Grygiel, R., Szmyt, M., & Evershed, R. P. (2013). Earliest evidence for cheese making in the sixth millennium bc in northern Europe. *Nature,* 493(7433), 522–525. https://doi.org/10.1038/nature11698

Salque, M., Regert, M., Evershed, R. P., Outram, A. K., Cramp, L. J. E., Decavallas, O., … Zoughlami, J. (2015). Widespread exploitation of the honeybee by early Neolithic farmers. *Nature*, 527(7577), 226–230. https://doi.org/10.1038/nature15757

Salque, M., Dunne, J., Altoft, D. T., Casanova, E., Cramp, L. J. E., Smyth, J., … Evershed, R. P. (2016). From the inside out: Upscaling organic residue analyses of archaeological ceramics. *Journal of Archaeological Science: Reports*. https://doi.org/10.1016/j.jasrep.2016.04.005

Saña, M. (2000). La gestió i explotació dels recursos animals. In A. Bosch, J. Chincilla, & J. Tarrís (Eds.), *El poblat lacustre neolític de la Draga. Excavacions de 1990 a 1998* (pp. 150–165). Banyoles: Monografies del CASC, 2. Museu d'Arqueologia de Catalunya.

Saña, M. (2011). La gestió dels recursos animals. In A. Bosch, J. Chinchilla, & J. Tarrís (Eds.), *El poblat lacustre neolític de la Draga. Excavacions 2000-2005* (pp. 117–212). Banyoles: Monografies del CASC, 9. Museu d'Arqueologia de Catalunya.

Saña, M. (2013). Domestication of animals in the Iberian Peninsula. In *The origins and spread of domestic animals in Southwest Asia and Europe* (pp. 195–220). Routledge.

Saña, M, & Navarrete, V. (2016). Gestió ramadera al pla de Barcelona durant la Prehistòria. In A Gómez & M. Molist (Eds.), *La prehistòria al pla de Barcelona* (pp. 97–103). Barcelona, Spain: Ajuntament de Barcelona.

Saña, M, Antolín, F., Bergadà, M., Castells, L., Craig, O., Edo, M., & Spiteri, C. (2015). Prácticas agropecuarias durante el Neolítico antiguo y medio en la cueva de Can Sadurní (Begues, Barcelona). In V. Gonçalves, M. Diniz, & A. C. Sousa (Eds.), *V Congreso do Neolítico Peninsular* (pp. 57–66). Lisboa: Centro de arqueologia da Universidade de Lisboa.

Saña, Maria, Tornero, C., & Nadal, J. (in press). *Gestió animal a les terres de l'Ebre entre 12000 cal aC i 4000 cal aC. In Història de les Terres de l'Ebre.* Amposta: Museu d'Amposta.

Sánchez, A., & Cañabate, M. L. (1998). I*ndicadores químicos para la arqueología.* Jaén: Servicio de Publicaciones de la Universidad de Jaén.

Sánchez, A., Cañabate, M. L., & Lizcano, R. (1998). Archaeological and chemical research on sediments and ceramics at polideportivo (spain): An integrated approach. *Archaeometry*, 40(2), 341–350. https://doi.org/10.1111/j.1475-4754.1998.tb00842.x

Sánchez, A., Parras, D., & Ramos, N. (2011). Análisis químico de contenidos en recipientes cerámicos. In C. Rueda (Ed.), *Territorio, culto e iconografia en los santuarios iberos del Alto Guadalquivir* (ss. IV a.n.e - I d.n.e).2 (1a ed., pp. 221–226). Jaén: CAA Ibérica. Universidad de Jaén.

Sanfeliu, D., & Flors, E. (2009). estudio tipológico y estilístico de la cerámica neolítica de costamar. In *Torre La Sal (Ribera de Cabanes, Castellón). Evolución Del Paisaje Antrópico Desde La Prehistoria Hasta El Medioevo.* Monografies de Prehistòria i Arqueologia Castellonenques, 8, 271–300.

Saul, H., Madella, M., Fischer, A., Glykou, A., Hartz, S., & Craig, O. E. (2013). Phytoliths in Pottery Reveal the Use of Spice in European Prehistoric Cuisine. *PLoS ONE*, 8(8), e70583. https://doi.org/10.1371/journal.pone.0070583

Schmidt, R., & Bancroft, I. (2011). *Genetics and Genomics of the Brassicaceae*. Springer.

Sebedio, J. L., LeQuere, J. L., Morin, O., Vatele, J. M., & Grandgirard, A. (1989). Heat Treatment of Vegetable Oils III. GC-MS Characterization of Cyclic Fatty Acid Monomers in Heated Sunflower and Linseed Oils After Total Hydrogenation. *Journal of the American Oil Chemists' Society*, 66(5), 704–709.

Ségurel, L., & Bon, C. (2017). On the Evolution of Lactase Persistence in Humans. *Annual Review of Genomics and Human Genetics*, 18(1), 297–319. https://doi.org/10.1146/annurev-genom-091416-035340

Seitz, L. M., & Paukstelis, J. V. (1977). Metabolites of Alternaria alternata: ergosterol and ergosta-4,6,8(14),22-tetraen-3-one. *Journal of Agricultural and Food Chemistry*, 25(4), 838–841. https://doi.org/10.1021/jf60212a030

Shackley, M. (1982). Gas chromatographic identification of a resinous deposit from a 6th century storage jar and its possible identification. *Journal of Archaeological Science*, 9(3), 305–306. https://doi.org/http://dx.doi.org/10.1016/0305-4403(82)90026-7

Sherratt, A. (1997). *Economy and society in prehistoric Europe: changing perspectives.* Edinburgh: Edinburgh University Press.

Sherratt, A. (1981). Plough and pastoralism: aspects of the secondary products revolution. In D. L. Clarke, I. Hodder, G. L. Isaac, & N. Hammond (Eds.), *Pattern of the past: studies in honour of David Clarke. Cambridge*: Cambridge Univeristy Press.

Sherriff, B. L., Tisdale, M. A., Sayer, B. G., Schwarcz, H. P., & Knyf, M. (1995). Nuclear magnetic resonance spectroscopic and isotopic analysis of carbonized residues from subarctic Canadian prehistoric pottery. *Archaeometry*, 37(1), 95–111. https://doi.org/10.1111/j.1475-4754.1995.tb00729.x

Shillito, L.-M. (2019). Building Stonehenge? An alternative interpretation of lipid residues in Neolithic Grooved Ware from Durrington Walls. *Antiquity*, 93(370), 1052–1060.

Shoda, S., Lucquin, A., Ahn, J., Hwang, C., & Craig, O. E. (2017). Pottery use by early Holocene hunter-gatherers of the Korean peninsula closely linked with the exploitation of marine resources. *Quaternary Science Reviews*, 170, 164–173. https://doi.org/10.1016/j.quascirev.2017.06.032

Skibo, J. M. (2013). *Understanding pottery function*. New York: Springer.

Smyth, J., & Evershed, R. P. (2014). Pottery , Archaeology and Chemistry Contents and Context. In A. Whittle & P. Bickle (Eds.), *Early farmers. The View from Archaeology and Science* (1st ed., pp. 347–367). Oxford: Proceedings of the British Academy, 198.

Šoberl, L., Horvat, M., Žibrat Gašparič, A., Sraka, M., Evershed, R., & Budja, M. (2014). Neolithic and Eneolithic activities inferred from organic residue analysis of pottery from Mala Triglavca, Moverna vas and Ajdovska jama, Slovenia. *Documenta Praehistorica*, 41(1), 149. https://doi.org/10.4312/dp.41.9

Solazzo, C., & Erhardt, D. (2007). Analysis of lipid residues in archaeological artifacts: marine mammal oil and cooking practices in the Arctic. In *Theory and practice of archaeological residue analysis* (Vol. British Ar, pp. 161–178). Oxford, UK: Archaeopress.

Soler, J., & Serangeli, J. (2007). *Memòria d'intervencions al Cau de les Guilles (Roses, Alt Empordà)*. Girona. Unpublished report.

Soler, J., & Serangeli, J. (2008). Internvencions arqueològiques de 2006 i 2007 al Cau de les Guilles de Roses. In J. Soler (Ed.), *Novenes Jornades d'Arqueologia de les Comarques de Girona* (pp. 57–62). Girona, Spain: MAC, L'Escala-Empúries.

Soler, J., & Serangeli, J. (2010). Les ocupacions al Cau de les Guilles de Roses des del Magdalenià fins a l'edat del bronze. *Cypsela*, 18, 79–92.

Sonneveld, W., Begemann, P. H., Van Beers, G. J., Keuning, R. van, & Schogt, J. C. M. (1962). 3, 7, 11, 15-Tetramethylhexadecanoic acid, a constituent of butterfat. *Journal of Lipid Research*, 3(3), 351–355.

Soriguer, M. C., Zabala, C., & Hernando, J. A. (2002). Características biológicas de la fauna marina del yacimiento de "El Retamar." In J. Ramos & M. Lazarich (Eds.), *Memoria de la excavación arqueológica en el asentamiento del VIo milenio a.n.a de "El Retamar"* (Puerto Real, Cádiz). (pp. 93–99). Spain: Junta de Andalucía, Consejeria de Cultura.

Sornoza, L., Barnolas, A., Arasa, A., Maestro, A., Rees, J. G., & Hernandez-molina, F. J. (1998). Architectural stacking patterns of the Ebro delta controlled by Holocene high-frequency eustatic fluctuations , delta-lobe switching and subsidence processes. *Sedimentary Geology*, 117, 11–32. https://doi.org/doi.org/10.1016/S0037-0738(97)00121-8

Spangenberg, J. E., Jacomet, S., & Schibler, J. (2006). Chemical analyses of organic residues in archaeological pottery from Arbon Bleiche 3, Switzerland – evidence for dairying in the late Neolithic. *Journal of Archaeological Science*, 33(1), 1–13. https://doi.org/10.1016/j.jas.2005.05.013

Spangenberg, J. E., & Ogrinc, N. (2001). Authentication of Vegetable Oils by Bulk and Molecular Carbon Isotope Analyses with Emphasis on Olive Oil and Pumpkin Seed Oil. *Journal of Agricultural and Food Chemistry*, 49(3), 1534–1540. https://doi.org/10.1021/jf001291y

Spiteri, C. (2012). *Pottery use at the transition to agriculture in the western Mediterranean. Evidence from biomolecular and isotopic caracterisation of organic residues in Impressed/Cardial Ware vessels.* PhD Thesis. Department of Archaeology. University of York.

Spiteri, C., Gillis, R. E., Roffet-Salque, M., Castells Navarro, L., Guilaine, J., Manen, C., ... Evershed, R. P. (2016). Regional asynchronicity in dairy production and processing in early farming communities of the northern Mediterranean. *Proceedings of the National Academy of Sciences*, 113(48), 13594–13599. https://doi.org/10.1073/pnas.1607810113

Steele, V. J., & Stern, B. (2017). Red Lustrous Wheelmade ware: Analysis of organic residues in Late Bronze Age trade and storage vessels from the eastern Mediterranean. *Journal of Archaeological Science: Reports*, (June), 0–17. https://doi.org/10.1016/j.jasrep.2017.06.027

Steele, V. J., Stern, B., & Stott, A. W. (2010). Olive oil or lard?: Distinguishing plant oils from animal fats in the archeological record of the eastern Mediterranean using gas chromatography/combustion/isotope ratio mass spectrometry. *Rapid Communications in Mass Spectrometry*, 24(23), 3478–3484. https://doi.org/10.1002/rcm.4790

Stern, B., Heron, C., Corr, L., Serpico, M., & Bourriau, J. (2003). Compositional Variations in Aged and Heated Pistacia Resin Found in Late Bronze Age Canaanite Amphorae and Bowls From Amarna, Egypt. *Archaeometry*, 45(3), 457–469. https://doi.org/10.1111/1475-4754.00121

Stern, B., Heron, C., Serpico, M., & Bourriau, J. (2000). A comparison of methods for establishing fatty acid concentration gradients across potsherds: A case study using late bronze age canaanite amphorae. *Archaeometry*, 42(2), 399–414. https://doi.org/10.1111/j.1475-4754.2000.tb00890.x

Stock, B. C., Jackson, A. L., Ward, E. J., Parnell, A. C., Phillips, D. L., & Semmens, B. X. (2018). Analyzing mixing systems using a new generation of Bayesian tracer mixing models. *PeerJ*, 6, e5096. https://doi.org/10.7717/peerj.5096

Sverrisdóttir, O. Ó., Timpson, A., Toombs, J., Lecoeur, C., Froguel, P., Carretero, J. M., … Thomas, M. G. (2014). Direct estimates of natural selection in iberia indicate calcium absorption was not the only driver of lactase persistence in Europe. *Molecular Biology and Evolution*, 31(4), 975–983. https://doi.org/10.1093/molbev/msu049

T

Taché, K., & Craig, O. E. (2015). Cooperative harvesting of aquatic resources and the beginning of pottery production in north-eastern North America. *Antiquity*, 89(343), 177–190. https://doi.org/doi:10.15184/aqy.2014.36

Tagliacozzo, A. (1994). Economic changes between the Mesolithic and the Neolithic in the Grotta dell'Uzzo (Sicily, Italy). *Accordia Research Papers*, 5, 7–37.

Tarifa, N. (2015). *Estudi funcional de les ceràmiques de la Cova del Sardo (Boí, Lleida) (4800-2500 cal ANE): les anàlisis de residus orgànics.* Ma Dissertation. Department of Prehistory. Autonomous University of Barcelona (UAB), Bellaterra.

Tarifa, N., Clop, X., Rosell, A., Camalich, M. D., Comes, P., Martín, D., … Rodríguez, F. J. (2019). New insights from Neolithic pottery analyses reveal subsistence practices and pottery use in early farmers from Cueva de El Toro (Málaga, Spain). *Archaeological and Anthropological Sciences.* https://doi.org/10.1007/s12520-019-00874-9

Tarrús, J. (1981). El Neolític Mitjà a les comarques gironines. In G. Camps, M. Llongueras, R. Marcet, M. A. Petit, & J. Rovira (Eds.), *El Neolític a Catalunya. Taula rodona de Montserrat.* (1a ed., pp. 87–102). Tortosa, Spain: Publicacions de l'Abadia de Montserrat.

Ten, R. (1979). Aproximació a l'estudi del Neolític Antic Final al Vallès. *Pyrenae,* 15–16, 21–41.

Terrats, N. (2008). Les ocupacions prehistòriques i antigues del turó de can Filuà. *L'ordit, l'annuari Del CREM,* (2), 51–71.

Thissen, L., Özbal, H., Türkekul, A., Gerritsen, F., & Özbal, R. (2010). The land of milk? Approaching dietary preferences of Late Neolithic communities in NW Anatolia. *Leiden Journal of Pottery Studies*, 26, 157–172.

Tieszen, L. L., Boutton, T. W., Tesdahl, K. G., & Slade, N. A. (1983). Fractionation and turnover of stable carbon isotopes in animal tissues: implications for δ 13 C analysis of diet. *Oecologia*, 57(1–2), 32–37.

Tishkoff, S. A., Reed, F. A., Ranciaro, A., Voight, B. F., Babbitt, C. C., Silverman, J. S., … Deloukas, P. (2007). Convergent adaptation of human lactase persistence in Africa and Europe. *Nature Genetics*, 39(1), 31–40. https://doi.org/10.1038/ng1946

Tresserras, J. (1997a). *Procesado y preparacion de alimentos vegetales para consumo humano. Aportaciones del estudio de fitolitos, almidones y lipidos en yacimientos prehistoricos y protohistoricos del cuadrante NE De la peninsula iberica.* PhD Thesis. Universitat de Barcelona.

Tresserras, J. (1997b). Identificación de residuos de cerveza en ánforas ibéricas de Alorda Park (Calafell, Baix Penedès) a través del estudio de indicadores microscópicos y bioquímicos. In: *Coloquio sobre tipologia y zonas de producción de ánforas ibéricas.* Madrid: Casa Velazquez y Universidad Autónoma de Madrid.

Tresserras, J. (1999). Estudio de residuos vegetales conservados en recipientes asociados a material de molienda en yacimientos de la Edad del Hierro del NE de la Península Ibérica. In R Buxó & E. Pons (Eds.), *Els productes alimentaris d'origen vegetal a l'edat del Ferro de l'Europa Occidental: de la producció al consum* (1a ed., pp. 371–377). Girona: Museu d'Arqueologia de Catalunya.

Tresserras, J. (2000a). La Cerveza: Un producto de consumo básico entre las comunidades ibéricas del N.E. peninsular. *SAGVNTUM*, 3(Extra-3), 139–145.

Tresserras, J. (2000b). Les resines vegetals. In A. Bosch, J. Chinchilla, & J. Tarrús (Eds.), *El poblat lacustre neolític de la Draga. Excavacions de 1990 a 1998* (p. 149). Monografies del CASC, 2. Museu d'Arqueologia de Catalunya.

Tresserras, J. (2004). Fuente Álamo (Almeria): Análisis de contenidos de recipientes cerámicos, sedimentos y colorantes procedentes de tumbas argáricas. *Madrider Mitteilungen*, 45, 132–139.

Tresserras, J. (2006). Análisis Arqueobotánico (fitolitos, almidones y fibras) y de residuos orgánicos. In P. Moret, J. A. Benavente, & A. Gorgues (Eds.), *Iberos del Matarraña* (pp. 287–291). Alcañiz: Al-Qannis: Taller de Arqueología de Alcañiz, 11.

Tresserras, J. (2009). Anàlisi de residus en ceràmica neolítica. Estudi del contingut d'un vas de boca quadrada de la mina 83. *Rubricatum*, 4(4), 93–95.

Tresserras, J., & Matamala, J. (2005). Estudio de residuos microscópicos y compuestos orgánicos en utillaje de molido y análisis de contenido de vasijas cerámicas. In P. Bueno, R. Balbín, & R. Barroso (Eds.), *El Dolmen de Azután (Toledo): áreas de habitación y areas funerarias en la cuenca interior del Tajo (pp. 235–241)*. Alcalá de Henares, Spain: Universidad de Toledo.

Tulloch, A. P. (1971). Beeswax: structure of the esters and their component hydroxy acids and diols. *Chem. Phys. Lipids*, 6(11684), 235–265.

Tulloch, A. P. (1973). Comparison of some commercial waxes by gas liquid chromatography. *Journal of the American Oil Chemists Society*, 50(9), 367–371. https://doi.org/10.1007/BF02640842

V

van Klinken, G. J., Richards, M. P., & Hedges, R. E. M. (2000). An Overview of Causes for Stable Isotopic Variations in Past European Human Populations: Environmental, Ecophysiological, and Cultural Effects. In S. H. Ambrose & M. A. Katzenberg (Eds.), *Biogeochemical approaches to paleodietary analysis.* (pp. 39–63). New York, USA: Kluwer Academic Publishers-Plenum Publishers.

Vicente, O., Gómez, A., Barcia, C., & Molist, M. (2014). Study of the spatial variability of Caserna de Sant Pau (Barcelona): old excavations, new approaches. In A. Garcia, J. Garcia, A. Maximiano, & J. Ríos-Garaizar (Eds.), *Debating Spatial Archaeology. Proceedings of the International Workshop on Landscape and Spatian Analysis in Archaeology* (pp. 267–276). Santander: Instituto Internacional de Investigaciones Prehistóricas de Cantabria.

Vidal, A. (2007). *Memòria científica de la intervenció arqueològica al jaciment de Mas d'en Boixos (Pacs del Penedès, Alt Penedès). Octubre de 2004–Febrer de 2005.* Arxiu del Servei d'Arqueologia i Paleontologia. Unpublished report.

Vika, E., & Theodoropoulou, T. (2012). Re-investigating fish consumption in Greek antiquity: Results from $\delta^{13}C$ and $\delta^{15}N$ analysis from fish bone collagen. *Journal of Archaeological Science*, 39(5), 1618–1627. https://doi.org/10.1016/j.jas.2012.01.016

Vilaseca, S. (1945). La cueva III de Les Quimeres, término municipal de Pradell (Tarragona). *Empúries: Revista de Món Clàssic i Antiguitat Tardana*, (7), 83–90.

Vilaseca, S., & Mercadé, J. (1956). Nueva localidad de cerámica cardial. *Butlletí Arqueològic*. Reial Societat Arqueològica Tarraconense, (53–56), 45–50.

Virella, X. (1973). Nous jaciments arqueològics a l'extrem nord-oest del massís del Garraf. *Vértex: Revista de La Federació Catalana de Muntanyisme*, 36, 422–425.

W

Washburn, D. K., Washburn, W. N., Shipkova, P. A., & Pelleymounter, M. A. (2014). Chemical analysis of cacao residues in archaeological ceramics from North America: considerations of contamination, sample size and systematic controls. *Journal of Archaeological Science*, 50(0), 191–207. https://doi.org/http://dx.doi.org/10.1016/j.jas.2014.07.011

Wattez, J. (1992). *Dynamique de formation des structures de combustion de la fin du Paléolithique au Neolithique moyen. Approche méthodologique et implications culturelles*. PhD Thesis. Université de Paris I.

Woodbury, S. E., Evershed, R. P., & Rossell, J. B. (1998). delta 13C analyses of vegetable oil fatty acid components, determined by gas chromatography--combustion--isotope ratio mass spectrometry, after saponification or regiospecific hydrolysis. *Journal of Chromatography. A*, 805(1–2), 249–257. Retrieved from http://www.ncbi.nlm.nih.gov/pubmed/9618921

Y

Yáñez, C., Burjachs, F., Tresserras, J., & Mestres, J. (2002). La fossa de Prats (Andorra), un jaciment del bronze mitjà al Pirineu. *Revista d'Arqueologia de Ponent*, 11–12, 123–150.

Yáñez, C., Malgosa, A., Burjachs, F., Díaz, N., García, C., Isidro, A., ... Matamala, J. (2002). El món funerari al final del V mil·leni a Andorra: la tomba de Segudet (Ordino). *Cypsela*, (14), 175–194.

Appendix 1

Appendix 1: PHP algorithm used to perform a posteriori additions

```
<HTML>
<HEAD>
<Title> A posteriori addition of sources</Title>
</HEAD>
<Body>
  <?PHP
$Dades  = array(“ProducteA”, “ProducteB”);
$Dades [“ProducteA”]= array (//insert here results from FRUITS//);
// Probabilistic data from case A

$Dades [“ProducteB”]=array(//insert here results from FRUITS//);
// Probabilistic data from case B

$g=1;
$i=1;
$j=1;
$x=1;
$llarg = count ($Dades [“ProducteB”]);
for ($g=1; $g <= $llarg ; $g++) {
    $i=$g-1;
  while ($i > 0) {
    $r=$i+$j;
    $pr[$x]= $r;
    $pa[$x]= $i;
    $pb[$x]= $j;
    $i--;
    $j++;
    $x++;
  }
  $i=1;
  $j=1;
}
// Calculation of all the possible additions between 1 and 100

echo(“<br>A posteriori addition<br>Total of instances:”);
echo count($pr);
echo (“<br><br>”);
$llarg= count($pr);
$j=0;
for ($i=2; $i <=$llarg ; $i++) {
  for ($x=0; $x<=$llarg ; $x++) {
    if ($pr[$x]==$i) {
      //echo ($pa[$x] .  “+” . $pb[$x] . “ = “ . $pr[$x] . “<br>”);
      $pbt = $pb[$x]-1;
      $pat = $pa[$x]-1;
```

```
      $Result[$j] = $Dades [“ProducteA”][$pat]*$Dades [“ProducteB”][$pbt];
      //echo ($Result[$j] .  “<br>”);
      $j++;
    }
  }
}
// Multiplications between product A and product B for all cases in each quantity.

$j=0;
$a=0;
$q=0;
$llarg2= count($Dades [“ProducteA”]);
for ($i=0; $i<=$llarg2 ; $i++) {
  for ($a=0; $a <=$q ; $a++) {
    $Estructura[$i][$a]=$Result[$j];
    $j++;
  }
  $q++;
}
// Orders the multiplication results to add them by quantity

echo (“<br><br>”);
$j=0;
$llarg3 = $llarg2-2;
$Final[$j] = 0;
for ($i=0; $i <=$llarg3 ; $i++) {
  $j++;
  $Final[$j] = array_sum ($Estructura[$i]);
}
// Adds the results of the multiplications

$llarg4 = count($Final);
$Total = array_sum($Final);
for ($i=0; $i <$llarg4 ; $i++) {
  $FinalN [$i] = $Final[$i]/$Total;
}
// Normalises the results

foreach ($FinalN as $i) {
echo (“<br>” . $i);
}
  ?>
</Body>
</HTML>
```

Appendix 2

Appendix 2

This database has been adapted from the one in Drieu 2017 annexe 4. It corresponds to available modern authentic reference δ^{13}C16:0 and δ^{13}C18:0 values.

ID	Species	Type	δ13C16:0‰	δ13C18:0 ‰	Δ13C‰	Geographic area	Bibliography
1	Ruminant	ruminant adipose	-29.6	-31.7	-2.1	UK	Craig et al., 2012
2	Cow	ruminant adipose	-29.1	-31.9	-2.8	UK	Copley et al., 2003 ; Dudd, 1999
3	Cow	ruminant adipose	-29.3	-32.15	-2.85	UK	Copley et al., 2003 ; Dudd, 1999
4	Cow	ruminant adipose	-30.25	-32	-1.75	UK	Copley et al., 2003 ; Dudd, 1999
5	Cow	ruminant adipose	-30.2	-32.7	-2.5	UK	Copley et al., 2003 ; Dudd, 1999
6	Sheep	ruminant adipose	-29.8	-32.2	-2.4	UK	Copley et al., 2003 ; Dudd, 1999
7	Sheep	ruminant adipose	-30.6	-32.7	-2.1	UK	Copley et al., 2003 ; Dudd, 1999
8	Sheep	ruminant adipose	-28.6	-30.4	-1.8	UK	Copley et al., 2003 ; Dudd, 1999
9	Sheep	ruminant adipose	-29.4	-30.8	-1.4	UK	Copley et al., 2003 ; Dudd, 1999
10	Sheep	ruminant adipose	-29.8	-31.6	-1.8	UK	Copley et al., 2003 ; Dudd, 1999
11	Sheep	ruminant adipose	-30.9	-32.9	-2	UK	Copley et al., 2003 ; Dudd, 1999
12	Sheep	ruminant adipose	-29.2	-30.5	-1.3	UK	Copley et al., 2003 ; Dudd, 1999
13	Sheep	ruminant adipose	-29.6	-31.5	-1.9	UK	Copley et al., 2003 ; Dudd, 1999
14	Sheep	ruminant adipose	-30.8	-32.6	-1.8	UK	Copley et al., 2003 ; Dudd, 1999
15	Sheep	ruminant adipose	-30.4	-32.2	-1.8	UK	Evershed et al., 1997
16	Sheep	ruminant adipose	-29.8	-31.8	-2	UK	Evershed et al., 1999
17	Sheep	ruminant adipose	-29.5	-31.9	-2.4	UK	Evershed et al., 1999
18	Sheep	ruminant adipose	-29.5	-32.3	-2.8	UK	Evershed et al., 1999
19	Ovine	ruminant adipose	-30.5	-33	-2.5		Craig et al., 2005
20	Cow	ruminant adipose	-28.3	-31.1	-2.8	UK	Evershed et al., 1997
21	Bovid	ruminant adipose	-28.8	-30.3	-1.5		Craig et al., 2005
22	White goat	ruminant adipose	-28.9	-31.3	-2.4	Switzerland, Grisons	Carrer et al., 2016
23	White goat	ruminant adipose	-30.2	-31.6	-1.5	Switzerland, Grisons	Carrer et al., 2016
24	Grey bull	ruminant adipose	-29.1	-32.1	-3	Switzerland, Grisons	Carrer et al., 2016
25	Cow	ruminant adipose	-30.25	-30.6	-0.35	Switzerland	Spangenberg et al., 2006
26	Cow	ruminant adipose	-31.65	-31.65	0	Switzerland	Spangenberg et al., 2006

27	Cow	ruminant adipose	-25.9	-25.8	0.1	Malta	Debono-Spiteri, 2012
28	Cow	ruminant adipose	-25.9	-26.1	-0.2	Malta	Debono-Spiteri, 2012
29	Sheep	ruminant adipose	-25.3	-27.8	-2.5	Malta	Debono-Spiteri, 2012
30	Sheep	ruminant adipose	-25.9	-27.3	-1.4	Malta	Debono-Spiteri, 2012
31	Goat	ruminant adipose	-26.3	-27.4	-1.1	Malta	Debono-Spiteri, 2012
32	Goat	ruminant adipose	-24.3	-26.3	-2	Malta	Debono-Spiteri, 2012
33	Cow	ruminant adipose	-22.8	-23.1	-0.3	Malta	Debono-Spiteri, 2012
34	Sheep	ruminant adipose	-22	-22.8	-0.8	Malta	Debono-Spiteri, 2012
35	Goat	ruminant adipose	-18.7	-19.3	-0.6	Malta	Debono-Spiteri, 2012
36	Sheep	ruminant adipose	-25.4	-25.28	0.12	Israel	Gregg et al., 2009
37	Goat	ruminant adipose	-31.16	-31.72	-0.56	Palestine	Gregg et al., 2009
38	Goat	ruminant adipose	-31.36	-31.6	-0.24	Palestine	Gregg et al., 2009
39		ruminant adipose	-29.64	-31.16	-1.52	Kenya	Dunne et al., 2012
40		ruminant adipose	-29.04	-30.58	-1.54	Kenya	Dunne et al., 2012
41		ruminant adipose	-29.2	-29.8	-0.6	Kenya	Dunne et al., 2012
42		ruminant adipose	-29.32	-29.8	-0.48	Kenya	Dunne et al., 2012
43		ruminant adipose	-27.6	-27.36	0.24	Kenya	Dunne et al., 2012
44		ruminant adipose	-27.24	-27.78	-0.54	Kenya	Dunne et al., 2012
45		ruminant adipose	-26.7	-27.8	-1.1	Kenya	Dunne et al., 2012
46		ruminant adipose	-26.5	-27.88	-1.38	Kenya	Dunne et al., 2012
47		ruminant adipose	-24.5	-25.3	-0.8	Kenya	Dunne et al., 2012
48		ruminant adipose	-24.14	-25.32	-1.18	Kenya	Dunne et al., 2012
49		ruminant adipose	-23.56	-25	-1.44	Kenya	Dunne et al., 2012
50		ruminant adipose	-21.06	-22.36	-1.3	Kenya	Dunne et al., 2012
51		ruminant adipose	-20.74	-21.94	-1.2	Kenya	Dunne et al., 2012
52		ruminant adipose	-20.72	-21.74	-1.02	Kenya	Dunne et al., 2012
53		ruminant adipose	-20.2	-22.4	-2.2	Kenya	Dunne et al., 2012
54		ruminant adipose	-18.8	-20.64	-1.84	Kenya	Dunne et al., 2012

55		ruminant adipose	-18.2	-20.76	-2.56	Kenya	Dunne et al., 2012
56		ruminant adipose	-17.76	-21	-3.24	Kenya	Dunne et al., 2012
57		ruminant adipose	-17.8	-19.28	-1.48	Kenya	Dunne et al., 2012
58		ruminant adipose	-17.1	-18.9	-1.8	Kenya	Dunne et al., 2012
59		ruminant adipose	-16.72	-19.6	-2.88	Kenya	Dunne et al., 2012
60		ruminant adipose	-16.28	-19.7	-3.42	Kenya	Dunne et al., 2012
61		ruminant adipose	-16.6	-19.2	-2.6	Kenya	Dunne et al., 2012
62		ruminant adipose	-16.5	-19.1	-2.6	Kenya	Dunne et al., 2012
63		ruminant adipose	-16.5	-18.52	-2.02	Kenya	Dunne et al., 2012
64		ruminant adipose	-16.5	-18	-1.5	Kenya	Dunne et al., 2012
65		ruminant adipose	-15.66	-19.1	-3.44	Kenya	Dunne et al., 2012
66		ruminant adipose	-15.66	-18.14	-2.48	Kenya	Dunne et al., 2012
67		ruminant adipose	-15.2	-18.04	-2.84	Kenya	Dunne et al., 2012
68	Pig	non-ruminant adipose	-25.9	-24.6	1.3	UK	Evershed et al., 1997
69	non-ruminant	non-ruminant adipose	-26.9	-25.2	1.7	UK	Copley et al., 2003
70	non-ruminant	non-ruminant adipose	-26.8	-25.2	1.6	UK	Copley et al., 2003
71	non-ruminant	non-ruminant adipose	-26.1	-24.8	1.3	UK	Copley et al., 2003
72	non-ruminant	non-ruminant adipose	-25.8	-24.5	1.3	UK	Copley et al., 2003
73	non-ruminant	non-ruminant adipose	-25.5	-24.15	1.35	UK	Copley et al., 2003
74	non-ruminant	non-ruminant adipose	-26.4	-25.4	1	UK	Copley et al., 2003
75	non-ruminant	non-ruminant adipose	-25.9	-24.8	1.1	UK	Copley et al., 2003
76	non-ruminant	non-ruminant adipose	-25.2	-24.4	0.8	UK	Copley et al., 2003
77	porcine	non-ruminant adipose	-26.8	-24.6	2.2		Craig et al., 2005

78	Pig	non-ruminant adipose	-24.7	-24.5	0.2	Malta	Spiteri 2012
79	Pig	non-ruminant adipose	-25	-25	0	Malta	Spiteri 2012
80	Pig	non-ruminant adipose	-29	-27.8	1.2	Switzerland	Spangenberg et al., 2006
81	Pig	non-ruminant adipose	-26	-26.9	-0.9	Switzerland	Spangenberg et al., 2006
82	Pig	non-ruminant adipose	-26.8	-25.8	1	UK	Colonese et al., 2017
83	Pig	non-ruminant adipose	-28.3	-27.2	1.1	UK	Colonese et al., 2017
84	Pig	non-ruminant adipose	-27.5	-26.6	0.9	UK	Colonese et al., 2017
85	Pig	non-ruminant adipose	-27.8	-27.2	0.6	UK	Colonese et al., 2017
86	Pig	non-ruminant adipose	-28.3	-27.2	1.1	UK	Colonese et al., 2017
87	Goose	non-ruminant adipose	-29.1	-27.7	1.4	UK	Dudd, 1999
88	Goose	non-ruminant adipose	-29.1	-28.4	0.7	UK	Dudd, 1999
89	Goose	non-ruminant adipose	-29.1	-28.2	0.9	UK	Dudd, 1999
90	Goose	non-ruminant adipose	-30.1	-28.5	1.6	UK	Dudd, 1999
91	Chicken	non-ruminant adipose	-29.3	-28.6	0.7	UK	Dudd, 1999
92	Chicken	non-ruminant adipose	-29.1	-28	1.1	UK	Dudd, 1999
93	Chicken	non-ruminant adipose	-29	-28.3	0.7	UK	Dudd, 1999
94	Chicken	non-ruminant adipose	-28.9	-28.3	0.6	UK	Dudd, 1999
95	Chicken	non-ruminant adipose	-29	-28.6	0.4	UK	Dudd, 1999
96	Chicken	non-ruminant adipose	-28.9	-28.2	0.7	UK	Dudd, 1999
97	Chicken	non-ruminant adipose	-29.4	-27.9	1.5	UK	Dudd, 1999

98	Chicken	non-ruminant adipose	-28.4	-27.7	0.7	UK	Dudd, 1999
99	Chicken	non-ruminant adipose	-28.6	-28.7	-0.1	UK	Colonese et al., 2017
100	Chicken	non-ruminant adipose	-28.7	-28.7	0	UK	Colonese et al., 2017
101	Chicken	non-ruminant adipose	-26.5	-26.2	0.3	UK	Colonese et al., 2017
102	Chicken	non-ruminant adipose	-26.1	-25.3	0.8	UK	Colonese et al., 2017
103	Chicken	non-ruminant adipose	-28.7	-28.8	-0.1	UK	Colonese et al., 2017
104	Chicken	non-ruminant adipose	-27	-26.7	0.3	UK	Colonese et al., 2017
105	Chicken	non-ruminant adipose	-27.1	-26.6	0.5	UK	Colonese et al., 2017
106	Chicken	non-ruminant adipose	-27.3	-27.1	0.2	UK	Colonese et al., 2017
107	Chicken	non-ruminant adipose	-28.8	-28.4	0.4	UK	Colonese et al., 2017
108	Chicken	non-ruminant adipose	-26.8	-26.8	0	UK	Colonese et al., 2017
109	Chicken	non-ruminant adipose	-27.3	-27.5	-0.2	UK	Colonese et al., 2017
110	Chicken	non-ruminant adipose	-27.6	-27.8	-0.2	UK	Colonese et al., 2017
111	Chicken	non-ruminant adipose	-27.6	-27.7	-0.1	UK	Colonese et al., 2017
112	Chicken	non-ruminant adipose	-27.9	-27.9	0	UK	Colonese et al., 2017
113	Chicken	non-ruminant adipose	-27.7	-27.9	-0.2	UK	Colonese et al., 2017
114	Horse	non-ruminant adipose	-30.1	-29.8	0.3	UK	Dudd, 1999
115	Horse	non-ruminant adipose	-31	-30.5	0.5	UK	Dudd, 1999
116	Horse	non-ruminant adipose	-30.9	-30	0.9	UK	Dudd, 1999
117	Horse	non-ruminant adipose	-30.4	-29.6	0.8	UK	Dudd, 1999

118	Horse	non-ruminant adipose	-30.7	-30.3	0.4	UK	Dudd, 1999
119	Horse	non-ruminant adipose	-29.9	-30	-0.1	UK	Dudd, 1999
120	Horse	non-ruminant adipose	-30	-27.9	2.1	UK	Dudd, 1999
121	Horse	non-ruminant adipose	-30.3	-30.1	0.2	UK	Dudd, 1999
122	Calf	young ruminant adipose	-27.45	-28.95	-1.5	Switzerland	Spangenberg et al., 2006
123	Calf	young ruminant adipose	-28.15	-28.6	-0.45	Switzerland	Spangenberg et al., 2006
124	Lamb	young ruminant adipose	-28	-26.7	1.3	Switzerland	Spangenberg et al., 2006
125	Lamb	young ruminant adipose	-28.8	-31.7	-2.9	Hébrides (UK)	Dudd, 1999
126	Lamb	young ruminant adipose	-29.1	-30.8	-1.7	UK	Copley et al., 2003 ; Dudd, 2009
127	Lamb	young ruminant adipose	-28.8	-30.5	-1.7	UK	Copley et al., 2003 ; Dudd, 2009
128	Roe deer	wild	-27.8	-31.6	-3.8	Germany	Debono-Spiteri, 2012
129	Roe deer	wild	-25.7	-26.9	-1.2	Germany	Debono-Spiteri, 2012
130	Roe deer	wild	-29.6	-30.7	-1.2	Switzerland, Grisons	Carrer et al., 2016
131	Chamois	wild	-28.4	-29.2	-0.8	Switzerland, Grisons	Carrer et al., 2016
132	Chamois	wild	-27.8	-30.2	-2.4	Switzerland, Grisons	Carrer et al., 2016
133	Ibex	wild	-28.1	-31	-2.8	Switzerland, Grisons	Carrer et al., 2016
134	Red deer	wild	-29.7	-34	-4.2	Switzerland, Grisons	Carrer et al., 2016
135	Deer	wild	-31.28	-33.45	-4.2		Spangenberg et al., 2006
136	Deer	wild	-32.5	-34.1	-1.6	UK	Lucquin et al., 2016b
137	Deer	wild	-32.4	-34.5	-2.1	UK	Lucquin et al., 2016b
138	Deer	wild	-28.7	-30.1	-1.4	UK	Dudd, 1999
139	Deer	wild	-29.2	-30.9	-1.7	UK	Dudd, 1999
140	Deer	wild	-29.8	-30.1	-0.3	UK	Dudd, 1999
141	Deer	wild	-29.2	-32.5	-3.3	UK	Dudd, 1999
142	Deer	wild	-28.4	-33.6	-5.2	UK	Dudd, 1999
143	Deer	wild	-31.1	-34.2	-3.1	UK	Dudd, 1999
144	Deer	wild	-31.2	-34	-2.8	UK	Dudd, 1999
145	Deer	wild	-29.7	-32.2	-4.2	UK	Craig et al., 2012
146	Red deer, bone	wild	-28.13	-31.89	-3.76	Poland	Craig et al., 2012

147	Red deer, bone	wild	-27.77	-31.54	-3.77	Poland	Craig et al., 2012
148	Red deer, bone	wild	-28.78	-32.98	-4.2	Poland	Craig et al., 2012
149	Red deer, bone	wild	-30.41	-34.06	-3.65	Poland	Craig et al., 2012
150	Red deer, bone	wild	-29.49	-33.08	-3.59	Poland	Craig et al., 2012
151	Red deer, bone	wild	-29.16	-33.43	-4.27	Poland	Craig et al., 2012
152	Red deer, bone	wild	-30.75	-33.44	-2.69	Poland	Craig et al., 2012
153	Red deer, bone	wild	-29.88	-33.47	-3.59	Poland	Craig et al., 2012
154	Red deer, bone	wild	-29.31	-32.69	-3.38	Poland	Craig et al., 2012
155	Red deer, bone	wild	-29.82	-33.41	-3.59	Poland	Craig et al., 2012
156	Deer	wild	-29	-32.4	-3.4	Japan	Horiuchi et al., 2015
157	Deer	wild	-24.7	-26.6	-1.9	Japan	Horiuchi et al., 2015
158	Moose, bone	wild	-29.12	-32.62	-3.5	Canada	Taché & Craig, 2015
159	Moose, bone	wild	-30.27	-32.8	-2.53	Canada	Taché & Craig, 2015
160	Moose, tissue	wild	-29.3	-32.21	-2.91	Canada	Taché & Craig, 2015
161	Moose, muscle	wild	-31.4	-32.5	-1.1	Alaska	Choy et al., 2016
162	Reindeer, bone	wild	-25.4	-28.32	-2.92	Canada	Taché & Craig, 2015
163	Reindeer, tissu	wild	-24.87	-26.14	-1.27	Canada	Taché & Craig, 2015
164	Boar	wild	-30.19	-28.53	1.66	Israel	Gregg et al., 2009
165	Boar	wild	-33.19	-31.48	1.71	Israel	Gregg et al., 2009
166	Boar	wild	-30.14	-28.03	2.11	Israel	Gregg et al., 2009
167	Boar	wild	-31.56	-27.51	4.05	Israel	Gregg et al., 2009
168	Boar	wild	-28.9	-28.8	0.1	Italy	Spiteri 2012
169	Boar	wild	-25.9	-25.6	0.3	Germany	Spiteri 2012
170	Boar	wild	-28.4	-27.1	1.3	Japan	Horiuchi et al., 2015
171	Boar	wild	-26.4	-26.9	-0.5	Japan	Horiuchi et al., 2015
172	Boar	wild	-28.1	-27.4	0.7	Japan	Horiuchi et al., 2015
173	Boar	wild	-28.1	-28.9	-0.8	Japan	Lucquin et al., 2016b
174	Boar	wild	-27.8	-27.9	-0.1	Japan	Lucquin et al., 2016b
175	Boar	wild	-27.8	-29.2	-1.4	Japan	Lucquin et al., 2016b
176	Boar	wild	-29.5	-29.6	-0.1	Japan	Lucquin et al., 2016b
177	Boar	wild	-28.9	-29.5	-0.6	Japan	Lucquin et al., 2016b
178	Boar	wild	-28.3	-27.4	0.9	Japan	Lucquin et al., 2016b
179	Boar	wild	-28.3	-27.2	1.1	Japan	Lucquin et al., 2016b
180	Boar	wild	-28.7	-27.7	1	Japan	Lucquin et al., 2016b
181	Boar	wild	-28.6	-27.8	0.8	Japan	Lucquin et al., 2016b
182	Boar	wild	-28.3	-27.4	0.9	Japan	Lucquin et al., 2016b
183	Bear	wild	-26.73	-27	-0.27	Canada	Taché & Craig, 2015

184	Beaver	wild	-31.04	-31.2	-0.16	Canada	Taché & Craig, 2015
185	Hare	wild	-32.15	-32.54	-0.39	Canada	Taché & Craig, 2015
186	Hare, muscle	wild	-30	-29.6	0.4	Alaska	Choy et al., 2016
187	Badger	wild	-29.15	-28.65	0.5	Canada	Taché & Craig, 2015
188	Muskrat	wild	-33.58	-33.02	0.56	Canada	Taché & Craig, 2015
189	Otter	wild	-31.76	-33.34	-1.58	Canada	Taché & Craig, 2015
190	Red squirrel, muscle	wild	-26.8	-27.6	-0.8	Alaska	Choy et al., 2016
191	Marmot	wild	-30.4	-29.9	0.4	Switzerland, Grisons	Carrer et al., 2016
192		Milk	-29.2	-34	-4.8	UK	Craig et al., 2012
193	Cow	Milk	-30.15	-36.4	-6.25	North UK	Craig et al., 2005
194	Cow	Milk	-28.15	-34.74	-6.59	UK	Craig et al., 2005
195		Milk	-28.3	-33.55	-5.25	UK	Copley et al., 2003
196		Milk	-27.8	-32.6	-4.8	UK	Copley et al., 2003
197		Milk	-29.3	-34.1	-4.8	UK	Copley et al., 2003
198		Milk	-28.25	-32.4	-4.15	UK	Copley et al., 2003
199		Milk	-28.5	-34.4	-5.9	UK	Copley et al., 2003
200		Milk	-30	-35.3	-5.3	UK	Copley et al., 2003
201		Milk	-29.4	-33.6	-4.2	UK	Copley et al., 2003
202		Milk	-29.8	-34.2	-4.4	UK	Copley et al., 2003
203		Milk	-31.2	-34.8	-3.6	UK	Copley et al., 2003
204		Milk	-29	-34.5	-5.5	UK	Copley et al., 2003
205		Milk	-25.75	-29.85	-4.1	Kazakhstan	Outram et al., 2009
206		Milk	-25.83	-30.4	-4.57	Kazakhstan	Outram et al., 2009
207		Milk	-26.42	-32.8	-6.38	Kazakhstan	Outram et al., 2009
208		Milk	-27.65	-30.77	-3.12	Kazakhstan	Outram et al., 2009
209		Milk	-28.45	-31.07	-2.62	Kazakhstan	Outram et al., 2009
210	Cow	Milk	-15.48	-21.6	-6.12	Kenya	Dunne et al., 2012
211	Cow	Milk	-15.4	-21.7	-6.3	Kenya	Dunne et al., 2012
212	Cow	Milk	-15.6	-21.8	-6.2	Kenya	Dunne et al., 2012
213	Cow	Milk	-18.2	-23.46	-5.26	Kenya	Dunne et al., 2012
214	Cow	Milk	-18.1	-23.8	-5.7	Kenya	Dunne et al., 2012
215	Cow	Milk	-24.76	-29.4	-4.64	Kenya	Dunne et al., 2012
216	Cow	Milk	-25.32	-30	-4.68	Kenya	Dunne et al., 2012
217	Cow	Milk	-26.16	-29.12	-2.96	Kenya	Dunne et al., 2012
218	Cow	Milk	-26.52	-29.4	-2.88	Kenya	Dunne et al., 2012
219	Goat	Milk	-26.64	-30.34	-3.7	Lybia	Dunne et al., 2012
220	Goat	Milk	-27.22	-30.48	-3.26	Lybia	Dunne et al., 2012
221	Goat	Milk	-24.34	-30.4	-6.06	Lybia	Dunne et al., 2012
222	Goat	Milk	-24.38	-30.2	-5.82	Lybia	Dunne et al., 2012
223	Goat	Milk	-26.42	-31.5	-5.08	Lybia	Dunne et al., 2012
224	Goat	Milk	-26.34	-31.98	-5.64	Lybia	Dunne et al., 2012
225	Goat	Milk	-25.52	-31.6	-6.08	Lybia	Dunne et al., 2012
226	Goat	Milk	-27.5	-31.9	-4.4	Lybia	Dunne et al., 2012
227	Goat	Milk	-28.5	-32.8	-4.3	Lybia	Dunne et al., 2012
228	Sheep, butter	Milk	-29.86	-34.24	-4.38	Jordan	Gregg et al., 2009

229	Goat	Milk	-25.7	-28.9	-3.2	Malta	Debono-Spiteri 2012
230	Goat	Milk	-24.2	-27.2	-3	Malta	Debono-Spiteri 2012
231	Sheep	Milk	-25.2	-29.8	-4.6	Malta	Debono-Spiteri 2012
232	Sheep	Milk	-25	-29.3	-4.3	Malta	Debono-Spiteri 2012
233	Sheep, C4	Milk	-22.4	-26.7	-4.3	Malta	Debono-Spiteri 2012
234	Sheep, cheese	Milk	-23.3	-28.4	-5.1	Trani, Italy	Debono-Spiteri 2012
235	Sheep, cheese	Milk	-22.1	-27.5	-5.4	Matera, Italy	Debono-Spiteri 2012
236	Goat, C4	Milk	-20.1	-27.2	-7.1	Malta	Debono-Spiteri 2012
237	Cow	Milk	-24.7	-27.1	-2.4	Malta	Debono-Spiteri 2012
238	Cow	Milk	-24.1	-27.3	-3.2	Malta	Debono-Spiteri 2012
239	Cow	Milk	-28.2	-32.4	-4.2	UK	Dudd, 1999
240	Cow	Milk	-31.2	-34.8	-3.6	UK	Dudd, 1999
241	Cow	Milk	-28.5	-34.4	-5.9	UK	Dudd, 1999
242	Cow	Milk	-27.8	-32.6	-4.8	UK	Dudd, 1999
243	Cow	Milk	-29	-34.5	-5.5	UK	Dudd, 1999
244	Cow	Milk	-30	-35.3	-5.3	UK	Dudd, 1999
245	Cow	Milk	-28.3	-33.5	-5.2	UK	Dudd, 1999
246	Cow	Milk	-29.3	-34.1	-4.8	UK	Dudd, 1999
247	Cow	Milk	-25.3	-31.3	-6	Germany	Debono-Spiteri
248	Cow	Milk	-27.4	-30.2	-2.8	Switzerland	Spangenberg et al., 2006
249	Cow	Milk	-27.8	-31.05	-3.25	Switzerland	Spangenberg et al., 2006
250	Cow	Milk	-28.8	-31.33	-2.53	Switzerland	Spangenberg et al., 2006
251	Cow	Milk	-27.3	-31.33	-4.03	Switzerland	Spangenberg et al., 2006
252	Cow	Milk	-28.1	-32.03	-3.93	Switzerland	Spangenberg et al., 2006
253	Cow	Milk	-26	-32.48	-6.48	Switzerland	Spangenberg et al., 2006
254	Cow	Milk	-27.8	-33.13	-5.33	Switzerland	Spangenberg et al., 2006
255	Cow	Milk	-28.43	-33.18	-4.75	Switzerland	Spangenberg et al., 2006
256	Cow	Milk	-28.85	-33.76	-4.91	Switzerland	Spangenberg et al., 2006
257	Cow, cheese	Milk	-28.2	-33.5	-5.2	Switzerland, Grisons	Carrer et al., 2016
258	Cow, cheese	Milk	-27.65	-32.1	-4.45	Switzerland	Spangenberg et al., 2006
259	Goat, cheese	Milk	-25.4	-27.93	-2.53	Switzerland	Spangenberg et al., 2006
260	Goat, cheese	Milk	-25.2	-29.1	-3.9	Switzerland	Spangenberg et al., 2006
261	Goat, cheese	Milk	-27.6	-32.3	-4.8	Switzerland, Grisons	Carrer et al., 2016
262	Goat	Milk	-28.2	-34.1	-5.9	Switzerland, Grisons	Carrer et al., 2016
263	Goat	Milk	-28.64	-31.4	-2.76	Switzerland	Spangenberg et al., 2006
264	Goat	Milk	-25.9	-32.65	-6.75	Switzerland	Spangenberg et al., 2006
265	Goat	Milk	-26.59	-33.5	-6.91	Switzerland	Spangenberg et al., 2006
266	Sheep	Milk	-29.4	-33.8	-4.4	UK	Dudd, 1999
267	Sheep	Milk	-29.8	-34.2	-4.4	UK	Dudd, 1999
268	Sheep	Milk	-31.7	-37.25	-5.55	Switzerland	Spangenberg et al., 2006
269	Sheep	Milk	-31.6	-38.25	-6.65	Switzerland	Spangenberg et al., 2006
270	Sheep	Milk	-31.3	-38.55	-7.25	Switzerland	Spangenberg et al., 2006

271	Sheep	Milk	-32.28	-39.45	-7.17	Switzerland	Spangenberg et al., 2006
272	Sheep, cheese	Milk	-28.35	-33.7	-5.35	Switzerland	Spangenberg et al., 2006
273	Sheep, cheese	Milk	-29.39	-34.15	-4.76	Switzerland	Spangenberg et al., 2006
274	Sheep, cheese	Milk	-31.2	-33.4	-2.2	Switzerland	Spangenberg et al., 2006
275	Anchovy	Marine organism	-24.5	-25.9	-1.4	Italy	Debono-Spiteri, 2012
276	Anchovy	Marine organism	-24.3	-25.4	-1.1	Italy	Debono-Spiteri, 2012
277	Bream	Marine organism	-24.1	-23.5	0.6	Malta	Debono-Spiteri, 2012
278	Bream	Marine organism	-23.8	-24.3	-0.5	Malta	Debono-Spiteri, 2012
279	Mullet	Marine organism	-22.2	-23.1	-0.9	Malta	Debono-Spiteri, 2012
280	Red mullet	Marine organism	-24.1	-24.3	-0.2	Malta	Debono-Spiteri, 2012
281	Bream	Marine organism	-22.1	-21.77	0.33	Japan	Craig et al., 2013
282	Bream	Marine organism	-22.36	-22.19	0.17	Japan	Craig et al., 2013
283	Perch	Marine organism	-23.38	-22.76	0.62	Japan	Craig et al., 2013
284	Mullet	Marine organism	-21.6	-20.97	0.63	Japan	Craig et al., 2013
285	Zebra sea bream	Marine organism	-21.45	-21.09	0.36	Japan	Craig et al., 2013
286	Mackerel	Marine organism	-23.9	-23.6	0.3	Japan	Horiuchi et al., 2015
287	Mackerel, bone	Marine organism	-24	-25.3	-1.3	Japan	Horiuchi et al., 2015
288	Mackerel, tissue	Marine organism	-23.7	-22.6	1.1	Japan	Horiuchi et al., 2015
289	Mackerel	Marine organism	-25	-25.2	-0.2	Japan	Lucquin et al., 2016b
290	Turbot	Marine organism	-22.9	-21.2	1.7	Japan	Horiuchi et al., 2015
291	Seriola, skin	Marine organism	-24.6	-23.8	0.8	Japan	Horiuchi et al., 2015
292	Seriola, tissue	Marine organism	-24.9	-25.6	-0.7	Japan	Horiuchi et al., 2015
293	Seriola, bone	Marine organism	-24.6	-22.9	1.7	Japan	Horiuchi et al., 2015
294	Seriola, bone	Marine organism	-24.7	-23.7	1	Japan	Horiuchi et al., 2015
295	Cod	Marine organism	-24.4	-24	0.4	Japan	Lucquin et al., 2016b
296	Cod	Marine organism	-24.9	-26.2	-1.3	UK	Dudd, 1999
297	Cod	Marine organism	-22.73	-22.19	0.54	Danemark	Craig et al., 2011
298	Cod	Marine organism	-22.74	-24.12	-1.38	Danemark	Craig et al., 2011

299	Cod	Marine organism	-22.04	-24.45	-2.41	Danemark	Craig et al., 2011
300	Cod	Marine organism	-21.3	-21.7	-0.4	Danemark	Craig et al., 2011
301	Burbot	Marine organism	-19.43	-21.01	-1.58	Danemark	Craig et al., 2011
302	Burbot	Marine organism	-21.15	-21.34	-0.19	Danemark	Craig et al., 2011
303	limande	Marine organism	-18.51	-19.82	-1.31	Danemark	Craig et al., 2011
304	plie	Marine organism	-19.81	-21.54	-1.73	Danemark	Craig et al., 2011
305	plie	Marine organism	-18.85	-20.07	-1.22	Danemark	Craig et al., 2011
306	limande	Marine organism	-18.9	-20.1	-1.2	Danemark	Lucquin et al., 2016b
307	hareng	Marine organism	-23.2	-20.8	2.4	Danemark	Craig et al., 2011
308	maquereau	Marine organism	-24.8	-25.1	-0.3		Craig et al., 2005
309	Cod	Marine organism	-24.2	-23.8	0.4		Craig et al., 2005
310	anguille	Marine organism	-18.1	-15.4	2.7	UK	Lucquin et al., 2016b
311	Tuna	Marine organism	-25.3	-24.9	0.4	UK	Lucquin et al., 2016b
312	Tuna	Marine organism	-26.6	-25.1	1.5	UK	Lucquin et al., 2016b
313	Haddock	Marine organism	-26.2	-23.9	2.3	UK	Dudd, 1999
314	Plaice	Marine organism	-24.1	-23.5	0.6	UK	Dudd, 1999
315	Salmon, muscle	Marine organism	-28.8	-27.2	1.6	Alaska	Choy et al., 2016
316	Salmon, muscle	Marine organism	-28.4	-26.6	1.8	Alaska	Choy et al., 2016
317	Salmon, muscle	Marine organism	-27.8	-25.6	2.2	Alaska	Choy et al., 2016
318	Salmon, muscle	Marine organism	-26.8	-26	0.8	Alaska	Choy et al., 2016
319	Salmon, muscle	Marine organism	-26.8	-25.4	1.4	Alaska	Choy et al., 2016
320	Salmon, muscle	Marine organism	-25.7	-24.1	1.6	Alaska	Choy et al., 2016
321	Truout	Marine organism	-16.89	-17.89	-1	Danemark	Craig et al., 2011
322	Truout, tissue	Marine organism	-25	-23.3	1.7	Japan	Lucquin et al., 2016b
323	Truout, tissue	Marine organism	-24.3	-24.6	-0.3	Japan	Lucquin et al., 2016b
324	salmon, experimental pot	Marine organism	-28.6	-29.1	-0.5	Japan	Lucquin et al., 2016b
325	salmon, experimental pot	Marine organism	-26.6	-25.6	1	Japan	Lucquin et al., 2016b

326	Salmon, tissue	Marine organism	-25.8	-24.8	1	Japan	Lucquin et al., 2016b
327	Salmon, tissue	Marine organism	-24.6	-23.5	1.1	Japan	Lucquin et al., 2016b
328	Salmon, tissue	Marine organism	-26.7	-25.3	1.4	Japan	Lucquin et al., 2016b
329	Salmon, tissue	Marine organism	-25.5	-25.4	0.1	UK	Lucquin et al., 2016b
330	Salmon, experimental pot	Marine organism	-25.3	-25.2	0.1	UK	Lucquin et al., 2016b
331	Oyster	Marine organism	-23.9	-24.5	-0.6	UK	Lucquin et al., 2016b
332	Clam	Marine organism	-25.2	-23.7	1.5	Japan	Horiuchi et al., 2015
333	Ruditapes (clam)	Marine organism	-24.7	-23.7	1	Japan	Lucquin et al., 2016b
334	Squid	Marine organism	-24.4	-23.8	0.6	Japan	Horiuchi et al., 2015
335	Harbour porpoise	Marine organism	-19.4	-20	-0.6	Japan	Horiuchi et al., 2015
336	Whale	Marine organism	-24.9	-23	1.9	Japan	Horiuchi et al., 2015
337	Sea lion	Marine organism	-24.5	-21.6	2.9	Japan	Horiuchi et al., 2015
338	Whale	Marine organism	-23.3	-23.5	-0.2	Japan	Lucquin et al., 2016b
339	Whale	Marine organism	-22.6	-23	-0.4	Japan	Lucquin et al., 2016b
340	Seal	Marine organism	-20	-19.95	0.05	Danemark	Craig et al., 2011
341	Seal	Marine organism	-12.8	-14.26	-1.46	Danemark	Craig et al., 2011
342	Seal	Marine organism	-18.56	-20.21	-1.65	Danemark	Craig et al., 2011
343	Seal, bone	Marine organism	-22.98	-24.15	-1.17	Canada	Taché & Craig, 2015
344	Seal, bone	Marine organism	-22.16	-23.78	-1.62	Canada	Taché & Craig, 2015
345	Seal, bone	Marine organism	-24.59	-24.28	0.31	Canada	Taché & Craig, 2015
346	Salmon, charred residue	Freshwater organism	-28.01	-28.72	-0.71	Japan	Craig et al., 2013
347	Salmon, charred residue	Freshwater organism	-25.22	-26.79	-1.57	Japan	Craig et al., 2013
348	Salmon, charred residue	Freshwater organism	-26.01	-27.33	-1.32	Japan	Craig et al., 2013
349	Trout, charred residue	Freshwater organism	-26.74	-26.72	0.02	Japan	Craig et al., 2013
350	Trout, charred residue	Freshwater organism	-27.64	-27.88	-0.24	Japan	Craig et al., 2013

351	Trout, charred residue	Freshwater organism	-25.83	-26.24	-0.41	Japan	Craig et al., 2013
352	Catfish	Freshwater organism	-27.41	-27.43	-0.02	Canada	Taché & Craig, 2015
353	Catfish	Freshwater organism	-26.07	-25.39	0.68	Canada	Taché & Craig, 2015
354	Catfish	Freshwater organism	-26.71	-27.16	-0.45	Canada	Taché & Craig, 2015
355	Catfish	Freshwater organism	-26.15	-26.21	-0.06	Canada	Taché & Craig, 2015
356	Catfish	Freshwater organism	-24.75	-24.81	-0.06	Canada	Taché & Craig, 2015
357	Catfish	Freshwater organism	-26.04	-26.38	-0.34	Canada	Taché & Craig, 2015
358		Freshwater organism	-25	-27	-2		Craig et al., 2011
359		Freshwater organism	-28.5	-29.5	-1		Craig et al., 2011
360		Freshwater organism	-35.6	-35.8	-0.2		Craig et al., 2011
361		Freshwater organism	-37.8	-37.4	0.4		Craig et al., 2011
362	Eel	Freshwater organism	-28.96	-29.22	-0.26	Danemark	Craig et al., 2011
363	Pike	Freshwater organism	-35.59	-35.84	-0.25	Danemark	Craig et al., 2011
364	Pike	Freshwater organism	-28.8	-26.4	2.4	UK	Lucquin et al., 2016b
365	Pike, muscle	Freshwater organism	-33.5	-31.3	2.2	Alaska	Choy et al., 2016
366	Pike, muscle	Freshwater organism	-36.4	-36.2	0.2	Alaska	Choy et al., 2016
367	Pike, muscle	Freshwater organism	-36.6	-35.6	1	Alaska	Choy et al., 2016
368	Tench	Freshwater organism	-28.53	-29.6	-1.07	Danemark	Craig et al., 2011
369	Tench	Freshwater organism	-25.04	-27.14	-2.1	Danemark	Craig et al., 2011
370	Tench	Freshwater organism	-37.95	-37.27	0.68	Danemark	Craig et al., 2011
371	Perch	Freshwater organism	-31.6	-29	2.6	UK	Lucquin et al., 2016b
372	Frostfish	Freshwater organism	-34.38	-34.63	-0.25	Canada	Taché & Craig, 2015
373	Frostfish	Freshwater organism	-34.19	-33.95	0.24	Canada	Taché & Craig, 2015
374	Frostfish	Freshwater organism	-33.86	-33.41	0.45	Canada	Taché & Craig, 2015
375	Frostfish	Freshwater organism	-33.35	-33.28	0.07	Canada	Taché & Craig, 2015
376	Frostfish	Freshwater organism	-32.58	-33.1	-0.52	Canada	Taché & Craig, 2015
377	Loach, muscle	Freshwater organism	-27.5	-28.8	-1.3	Alaska	Choy et al., 2016

378	Loch, muscle	Freshwater organism	-30.4	-29.4	1	Alaska	Choy et al., 2016
379	Clam	Freshwater organism	-28.6	-28.1	0.5	Japan	Lucquin et al., 2016b
380	Clam	Freshwater organism	-30.3	-29.7	0.6	Japan	Horiuchi et al., 2015
381	Carp	Freshwater organism	-30.8	-28.8	2	UK	Lucquin et al., 2016b
382	Mussel	Freshwater organism	-34.6	-35	-0.4	Alaska	Choy et al., 2016
383	Mussel	Freshwater organism	-35.2	-34.9	0.3	Alaska	Choy et al., 2016
384	Mussel	Freshwater organism	-41.5	-39.6	1.9	Alaska	Choy et al., 2016
385		Freshwater organism	-31.4	-30.93	0.47	Kazakhstan	Outram et al., 2009
386		Freshwater organism	-31.6	-31.1	0.5	Kazakhstan	Outram et al., 2009
387		Freshwater organism	-31.3	-31.3	0	Kazakhstan	Outram et al., 2009
388		Freshwater organism	-31.8	-31.6	0.2	Kazakhstan	Outram et al., 2009
389		Freshwater organism	-32.2	-32.1	0.1	Kazakhstan	Outram et al., 2009
390		Freshwater organism	-32.8	-32.2	0.6	Kazakhstan	Outram et al., 2009
391		Freshwater organism	-32.46	-32.6	-0.14	Kazakhstan	Outram et al., 2009
392		Freshwater organism	-32.83	-32.9	-0.07	Kazakhstan	Outram et al., 2009
393	White fish	Freshwater organism	-33.3	-32.25	1.05		Spangenberg et al., 2006
394	White fish	Freshwater organism	-34.8	-33.8	1		Spangenberg et al., 2006
395	Amur minnow, charred residue	Freshwater organism	-27.43	-28.13	-0.7	Japan	Craig et al., 2013
396	Stone moroko, charred residue	Freshwater organism	-26.95	-26.64	0.31	Japan	Craig et al., 2013
397	olive	Plant	-33	-31.9	1.1	Slovenia	Spangenberg & Ogrinc, 2001
398	olive	Plant	-29.9	-29.8	0.1	Slovenia	Spangenberg & Ogrinc, 2001
399	olive	Plant	-30.7	-30.8	-0.1	Slovenia	Spangenberg & Ogrinc, 2001
400	olive	Plant	-31.4	-30.8	0.6	Slovenia	Spangenberg & Ogrinc, 2001
401	olive	Plant	-32.2			Slovenia	Spangenberg & Ogrinc, 2001
402	olive	Plant	-31	-31.4	-0.4	Slovénie	Spangenberg & Ogrinc, 2001
403	olive	Plant	-31.5	-31	0.5	Croatia	Spangenberg & Ogrinc, 2001

404	olive	Plant	-30.4	-30.6	-0.2	Croatia	Spangenberg & Ogrinc, 2001
405	olive	Plant	-29.1	-30	-0.9	Croatia	Spangenberg & Ogrinc, 2001
406	olive	Plant	-32	-31.3	0.7	Croatia	Spangenberg & Ogrinc, 2001
407	olive	Plant	-33.8			Croatia	Spangenberg & Ogrinc, 2001
408	olive	Plant	-30.4			Croatia	Spangenberg & Ogrinc, 2001
409	olive	Plant	-31	-29.7	1.3	Croatia	Spangenberg & Ogrinc, 2001
410	olive oil	Plant	-28.5	-28.5	0		Dudd, 1999
411	olive oil	Plant	-28.3	-27.5	0.8		Steele et al., 2010
412	olive oil	Plant	-28.25	-28.65	-0.4		Steele et al., 2010
413	olive oil	Plant	-28.35	-29	-0.65		Steele et al., 2010
414	almond oil	Plant	-27.65	-29.5	-1.85		Steele et al., 2010
415	Hazlenut oil	Plant	-27.35	-27.3	0.05		Steele et al., 2010
416	Walnut	Plant	-31.5	-31.4	0.1	Japan	Horiuchi et al., 2015
417	Chesnut	Plant	-35.1	-32.7	2.4	Japan	Horiuchi et al., 2015
418	White acorn	Plant	-34.7	-33.4	1.3	Japan	Horiuchi et al., 2015
419	Acorn	Plant	-37.6	-35.7	1.9	Japan	Horiuchi et al., 2015
420	Acorn	Plant	-33.7	-34.1	-0.4	Japan	Lucquin et al., 2016b
421	Acorn	Plant	-32.4	-33.3	-0.9	Japan	Lucquin et al., 2016b
422	Acorn	Plant	-34.8	-35.6	-0.8	Japan	Lucquin et al., 2016b
423	Acorn	Plant	-32.8	-34.3	-1.5	Japan	Lucquin et al., 2016b
424	Acorn	Plant	-31.8	-33.7	-1.9	Japan	Lucquin et al., 2016b
425	Pumpkin	Plant	-29.7	-31.2	-1.5	Slovenia	Spangenberg & Ogrinc, 2002
426	Pumpkin	Plant	-29.7	-27.3	2.4	Slovenia	Spangenberg & Ogrinc, 2002
427	Pumpkin	Plant	-31.8			Slovenia	Spangenberg & Ogrinc, 2002
428	Pumpkin	Plant	-31.8			Slovenia	Spangenberg & Ogrinc, 2002
429	Sunflower	Plant	-32	-31.2	0.8	Slovenia	Spangenberg & Ogrinc, 2003
430	Sunflower	Plant	-30.8	-29.6	1.2	Slovenia	Spangenberg & Ogrinc, 2004
431	Soy	Plant	-32.4	-32.5	-0.1		Spangenberg & Ogrinc, 2005
432	Sesame	Plant	-28.1	-27.9	0.2		Spangenberg & Ogrinc, 2005
433	Maize	Plant	-22.4			Slovenia	Spangenberg & Ogrinc, 2005
434	Rapeseed	Plant	-31.3			Slovenia	Spangenberg & Ogrinc, 2005
435	Date	Plant	-32	-31.9	0.1		Copley, 2002
436	Radish	Plant	-32.5	-32.1	0.4		Copley, 2002
437	Castor	Plant	-33.7	-32.9	0.8		Copley, 2002
438	Barley	Plant	-32.7				Copley, 2002

439	Wheat	Plant	-33.9			Copley, 2002
440	Sorghum	Plant	-18.4	-18.7	-0.3	Copley, 2002
441	Lablab	Plant	-34.3	-34.3	0	Copley, 2002
442	Gingerbread tree seed	Plant	-34.2	-34.1	0.1	Copley, 2002

Appendix 3

Data from Carrer Reina Amàlia

Lab id	Rim	Width	Diam	Apert	Cook	Crust	Decor	Weight
8090	Indet	11	-	Indet	M	0	Plain cord	254
8656	Indet	14	-	Indet	O	0	Plain cord	206
8890	F1	10	31.5	Straight	O	1	Plain cord	263
8900	H1	11	-	Closed	M	1	Plain cord	208
9086	Indet	13	-	Indet	M	0	Plain cord	219
9092	I1	15	-	Closed	R	1	T shaped Plain cord	205
9189	IF1	8	unkwn	Closed	M	1	Handle	240
9296	I9	11	-	Closed	M	0	Plain cord	196
9343	F1	15	unkwn	Straight	M	0	Plain cord	207
9409	E4	11	-	Open	R	0	Plain cord	205
9470	F4	14	-	Straight	R	0	Plain cord	200
9565	F1	10	-	Straight	M	0	Hande	201
9630	IF1	10	-	Closed	R	0	Handle	217
9674	I1	10	unkwn	Closed	R	0	None	202
9679	F1	12	-	Straight	R	1	Plain cord	194
9680	H1	8	-	Closed	M	0	Handle	208
9684	H9	9	20	Closed	M	1	Handle	212
9734	Indet	13	-	Indet	M	0	None	202
9359	IF1	12	-	Closed	R	1	Plain cord	206

Appendix 3. Table 1: Studied vessels from stratigraphic unit 95 (Phase I) in Carrer Reina Amàlia.

Lab id	Rim	Width	Diam	Apert	Cook	Crust	Decor
4892	Rim	11.0	45	Straight	R	1	No
5132	Rim	15.8	40	Open	R	0	Yes
5531	Rim	11.0	45	Open	R	1	Yes
5561	Rim	11.4	45	Straight	M	1	Yes
5823	Rim	12.4	50	Open	R	1	No
6100	Rim	11.9	45	Straight	R	1	Yes
6162	Rim	8.1	-	Straight	R	0	No
6231	Rim	6.0	25	Straight	R	1	No
6405	Rim	12.8	50	Straight	R	1	Yes
6522	Rim	10.0	45	Closed	M	1	Yes
6681	Rim	13.7	45	Open	M	1	No
6695	Rim	13.0	50	Closed	R	0	No
6725	Rim	11.8	50	Open	R	0	No
7656	Rim	11.5	35	Closed	M	1	Yes
7718	Rim	12.1	25	Straight	O	1	Yes
7778	Rim	11.9	50	Straight	M	1	Yes
8009	Rim	7.2	25	Open	M	1	No
8070	Rim	10.0	45	Open	R	1	No
8234	Rim	11.8	45	Straight	M	0	No
8263	Rim	12.4	50	Straight	R	0	Yes
8392	Base	13.0	-	-	R	1	No
8413	Rim	10.5	45	Straight	M	1	Yes
8439	Rim	12.3	45	Straight	M	0	No
8721	Rim	12.8	45	Open	M	0	Yes
8726	Rim	10.7	50	Straight	M	0	No
8737	Rim	9.4	20	Straight	R	1	No
8764	Rim	11.1	40	Closed	R	1	Yes
8842	Wall	11.8	-	-	O	1	No
8995	Rim	14.0	45	Open	R	0	Yes
9064	Rim	14.5	45	Straight	R	1	No
9065	Rim	12.4	20	Open	M	0	No
9454	Rim	7.6	30	Open	M	1	No
11960	Rim	10.3	40	Closed	R	0	No

Appendix 3. Table 2: Studied vessels from stratigraphic unit 59 (Phase II) in Carrer Reina Amàlia. Sample weight was roughly around 1 g.

Statistical results	**TLE**	**PS**	**Temperature**	**Phytanic acid**	**Isotopes**
Rim type	Kruskal-Wallis Hc=1.385 n=46 p=0.50	ANOVA F=3.06, n=43, p=0.0572	x2=0.86 df=2 n=43 p=0.65	x2=4.45 df=2 n=42 p=0.11	No correlation
Firing conditions	Krustal-Wallis Hc=6.246 n=52 p=0.044	ANOVA F=0.2482 n=52 p=0.781	x2=3.1682 df=2 n=52 p=0.205	x2=5.9639 df=2 n=52 p=0.051	No correlation
Crust presence	Mann-Withney U 282 n=52 p=0.326	Student's T=2.5442 n=52 p=0.014	x2=3.166 df=1 n=52 Fisher's exact p=0.123	x2=3.48 df=1 n=52 Fisher's exact p=0.085	No correlation
Decoration	Kruskal-Wallis Hc=1.079 n=52 p=0.58	ANOVA F=0.302 n=52 p=0.7355	x2=1.048 df=2 n=52 p=0.592	x2=3.09 df=2 n=52 p=0.212	No correlation
Width	Spearman's D 21350 n=52 p=0.535	Pearson's r=-0.177 n=52 p=0.207	Student's T=2.1475 n=52 p=0.0366	Student's T=0.418 n=52 p=0.677	Student's T=2.580 n=10 p=0.033

Appendix 3. Table 3: Statistical tests evaluating possible correlations between pottery and residue characteristics.

Lab ID	FFA range	C16:0	C18:0	C15:0	C17:0	C19:0	C15:0br	C17:0br
5561	14-18	42.5	34.9	2.8	1.0	0.0	0.0	0.0
8439	14-18	44.5	38.4	1.6	0.0	0.0	0.0	0.0
4892	14-18	42.3	23.7	4.1	1.1	0.0	1.1	0.0
8263	14-18	44.7	16.4	5.2	0.8	0.0	4.1	0.0
11960	14-18	40.8	32.1	3.1	1.0	0.0	0.0	0.0
7656	14-22	38.2	31.8	5.2	1.6	0.0	0.4	0.0
7718	14-18	36.7	50.0	2.0	0.5	0.0	0.5	0.0
8764	14-22	35.6	16.9	4.5	1.2	0.0	2.5	0.0
6162	13-30	32.3	37.9	2.1	2.4	0.8	0.3	0.3
8392	14-20	33.9	61.6	0.5	0.3	0.0	0.0	0.0
6522	14-20	41.5	49.3	1.2	0.3	0.0	0.0	0.0
8842	14-20	38.3	54.7	1.0	0.5	0.0	0.0	0.0
8009	13-24	28.7	47.3	4.7	5.8	0.6	0.4	0.4
9065	14-26	32.9	36.4	2.0	1.6	0.7	0.5	0.6
7778	14-20	38.1	46.0	1.4	1.3	0.1	0.0	0.2
8726	12-28	27.3	41.8	6.4	9.3	1.4	0.6	1.7
5823	14-28	29.2	51.8	0.9	3.2	1.1	0.3	0.9
6231	12-28	31.1	42.2	2.0	3.1	1.0	0.5	1.0
8721	12-26	36.0	41.6	2.7	4.9	0.8	0.6	1.5
9064	12-30	37.3	43.3	1.3	4.1	0.7	0.2	1.1
6681	12-28	27.9	52.2	1.0	4.4	1.3	0.1	1.1
6725	12-28	32.5	45.9	1.9	3.2	1.0	0.3	1.2
5531	12-30	32.2	43.1	2.1	5.4	1.0	0.4	1.7
5132	12-28	35.8	38.9	1.9	3.3	0.8	0.6	1.2
8413	12-26	30.2	47.4	1.5	4.1	0.8	0.3	1.5
6100	12-28	29.6	47.9	1.6	3.9	1.2	0.3	1.3
9464	11-28	28.9	37.5	1.9	5.1	1.1	0.5	2.5
8070	9-32	29.9	31.0	3.3	4.0	1.0	1.0	2.6
6405	11-28	34.4	32.3	6.2	7.4	1.1	0.7	2.3
6695	9-13	30.0	38.1	2.5	5.3	1.2	0.5	2.1
8737	12-27	30.2	42.5	2.1	5.0	1.3	0.5	2.6
8234	11-26	34.2	35.7	3.2	4.4	1.0	0.6	2.2
8995	11-28	35.5	38.3	2.6	5.4	1.1	0.5	2.5

Appendix 3. Table 4: Relative quantities of other free fatty acids and the range of free fatty acids detected in samples from stratigraphic unit 59 (Phase II) in Carrer Reina Amàlia.

Molecule	Retention time		Sherds containing the compound		Abundance as % of the total	
	DCM/MeOH	Acid Methanol	DCM/MeOH	Acid Methanol	DCM/MeOH	Acid Methanol
Glycerol	10.00		10%		0.00	
C9:0	11.00		10%		0.00	
C10:0	12.24		30%		0.01	
OH-C12	13.57		40%		0.12	
C12:0	14.51	13.01	60%	60%	0.33	0.1
C13:0	15.60	14.16	60%	60%	0.39	0.2
C14:1		15.03		10%		0.0
OH-C14	15.74		50%		0.15	
C14:0	16.60	15.28	60%	100%	3.74	3.1
C15:0br	17.22	15.93	30%	60%	0.11	0.2
C15:0br	17.30	16.02	20%	50%	0.06	0.2
C15:0br		16.07		10%		0.0
C15:0	17.58	16.33	60%	100%	2.59	2.2
C16:0 UND	17.63		60%		1.60	
C16 OH	17.72		40%		0.18	
C16:0br	18.17	16.96	30%	60%	0.15	0.4
C16:1		17.20		10%		0.0
C16:1	18.27	17.08	60%	100%	0.42	0.9
OH-C17	18.38		10%		0.01	
C16:0	18.56	17.36	70%	100%	34.46	31.5
C17:0br	19.08	17.94	60%	60%	0.82	0.9
C17:0br	19.16	18.02	60%	70%	0.62	0.7
C17:0	19.42	18.30	60%	100%	5.02	4.5
2-hydroxy C16:0		18.48		10%		0.0
C18 OH	19.53		40%		0.08	
C18:0 UND	19.68		60%		12.82	
C18:1	20.05	18.88	60%	30%	0.92	0.1
C18:1	20.11	18.98	10%	100%	0.13	2.4
C18:1	20.14	19.05	40%	60%	1.31	2.1
C18:0	20.32	19.26	60%	100%	31.68	43.9
2-hydroxy C18:0	20.84	20.88	20%	40%	0.13	0.1
C19:0br		19.78		40%		0.1
C19:0br		19.86		40%		0.1
C19:0	21.06	20.10	20%	70%	0.04	1.1
oxo-C18:0		20.55		50%		0.1
oxo-C18:0		20.74		60%		0.5
C20:0 UND	21.24		30%		0.08	
MAG	21.63		20%		0.12	
C20:0	21.89	20.96	30%	100%	0.03	2.3
oxo-C18:0		21.00		10%		0.0
MAG	22.39		10%		0.01	
MAG	22.87		30%		0.23	
MAG	23.12		50%		0.71	
C21:0		21.76		60%		0.4
C22:1		22.34		10%		0.0
C22:0	23.39	22.51	10%	60%	0.01	0.6

C23:0		23.17		60%		0.4
C24:0		23.91		60%		0.5
C25:0		24.62		60%		0.2
C26:0		25.42		60%		0.1
C27:0		26.08		50%		0.0
C28:0		26.71		50%		0.0
C30:0		27.92		10%		0.0
MAG	24.27		30%		0.10	
MAG	24.51		30%		0.49	
Squalene	24.72		60%			
Cholesterol	26.95		90%			
DAG	30.98		10%		0.00	
DAG	31.17		10%		0.00	

Appendix 3. Table 5: Retention times and percentage of sherds where the compound was present. Data from samples in UE59 (Phase II)in Carrer Reina Amàlia where both solvent and acidified methanol extractions were practiced.

Appendix 4

Appendix 4

Statistical tests supporting the discussion section

5500-4500	Non-Ruminant is present	Non-Ruminant is absent
Dairy is present	0	6
Dairy is absent	4	2

Appendix 4. Table 1: Contingency table for the 12 estudies sites where more than 1 sample was submitted to compound specific isotopic analyses.

5500-5000	Non-Ruminant is present	Non-Ruminant is absent
Dairy is present	0	5
Dairy is absent	4	0

Appendix 4. Table 2: Contingency table for the 9 studies from the Cardial Early Neolithic period where more than 1 sample was submitted to compound specific isotopic analyses.

Case	N	DF	χ^2	Fisher's exact P	Hypothesis
5500-4500	12	1	5.23	0.060606	No association (null) hypothesis should be kept
5500-5000	8	1	8	0.028571	No association (null) hypothesis should be rejected

Appendix 4. Table 3: Results of the χ^2 test for the data in tables 1 and 2.

Variable	N	Shapiro-Wilk	P(normal)	Hypothesis
Modelled Non-ruminant median	11	0.9155	0.2832	Normallity (null) hypothesis is kept
Modelled dairy median	11	0.8604	0.05831	Normallity (null) hypothesis is kept
Modelled non-ruminant median without Vidre and Font Major	9	0.9122	0.3314	Normallity (null) hypothesis is kept
Modelled dairy median withouth Vidre and Font Major	9	0.938	0.5614	Normallity (null) hypothesis is kept
Modelled non-ruminant medial Cardial	8	0.9157	0.3963	Normallity (null) hypothesis is kept
Modelled dairy medial Cardial	8	0.8614	0.1241	Normallity (null) hypothesis is kept

Appendix 4. Table 4: Normality tests for the modelled median quantity of non-ruminant and dairy products in sites from this PhD.

Case	N	Pearsons'r	P(uncorr)	Hypothesis
All cases	11	-0.54171	0.0076343	No correlation (null) hypothesis is rejected.
Removes Vidre and Font Major	9	-0.85707	0.0031515	No correlation (null) hypothesis is rejected.
Only Cardial	8	-0.74173	0.035158	No correlation (null) hypothesis is rejected.

Appendix 4. Table 5: Values resulting from testing the possible correlation between non-ruminant and dairy modelled inputs in sites from this PhD

Case	r^2	N	Shapiro-Wilk	P(normal)	Hypothesis
All cases	0.56	11	0.8899	0.1386	Normallity (null) hypothesis is kept
Removes Vidre and Font Major	0.73	9	0.9767	0.9449	Normallity (null) hypothesis is kept
Cardial	0.55	8	0.9276	0.4941	Normallity (null) hypothesis is kept

Appendix 4. Table 6: Study of the strenght of the correlation and the residual normality to further evaluate the detected correlations in table 5.

Variable	N	ShapiroWilk	P(normal)	Hypothesis
D-NR	11	0.9534	0.6872	Normallity (null) hypothesis is kept
m.a.s.l	11	0.8258	0.0259	Normallity (null) hypothesis is rejected
Distance from sea	11	0.8843	0.1178	Normallity (null) hypothesis is kept
Distance from sea <30	9	0.8313	0.0462	Normallity (null) hypothesis is rejected
Open air sites	7	0.937	0.6118	Normallity (null) hypothesis is kept
Cave sites	4	0.8772	0.327	Normallity (null) hypothesis is kept
Ovicaprine NR	7	0.9297	0.548	Normallity (null) hypothesis is kept
5500-5000 Ovicaprine NR	6	0.8775	0.258	Normallity (null) hypothesis is kept
Bovids NR	7	0.8216	0.066	Normallity (null) hypothesis is kept

Appendix 4. Table 7: Normality tests for variables characterising sites from this PhD.

Case	N	Test	Statistic	P	Hypothesis
Masl	11	Spearman's D	143	0.27246	No correlation (null) hypothesis is kept
Distance from sea	11	Perasons'r	0.23334	0.48986	No correlation (null) hypothesis is kept
Distance from sea <30	9	Spearman's D	0.77366	0.014439	No correlation (null) hypothesis is rejected
Open air/Caves	11	Student's T same variance	0.1902	0.85337	Same mean (null) hypothesis is kept
Ovicaprine NR	7	Perasons'r	0.70526	0.0767	No correlation (null) hypothesis is kept
5500-5000 Ovicaprine NR	6	Perasons'r	0.83966	0.03650	No correlation (null) hypothesis is rejected
Ovicaprines and Bovids in flocks	7	Perasons'r	0.94911	0.001	No correlation (null) hypothesis is rejected

Appendix 4. Table 8: Statistical test evaluating the correlation between the D-NR variable and several site characteristics.

Case	r^2	N	Shapiro-Wilk	P(normal)	Hypothesis
Distance from sea <30	0.59	9	0.8277	0.04207	Normallity (null) hypothesis is rejected
5500-5000 Ovicaprine NR	0.70	6	0.9611	0.8283	Normallity (null) hypothesis is kept
Ovicaprines and Bovids in flocks	0.87	7	0.9501	0.7309	Normallity (null) hypothesis is kept

Appendix 4. Table 9: Study of the strenght of the correlation and the residual normality to further evaluate the detected correlations in table 8.

Appendix 5

	Period	Region	Type	N	Rims	Walls	Bases	Appliquée	Impressed Decoration	Residues	Ruminant	Non Ruminant	Dairy	>250°C
CARDIAL				115										
Sant Pau	5600-5000	Barcelona	Open air	24	25%	75%	0	17%	8%	92%	57%	43%	No	No
Berenguer	5600-5400	Barcelona	Open air	3	0	100%	0	0	0	0	No	No	No	No
Guixeres Vilobí	5600-5400	G & P	Open air	22	59%	41%	0	41%	14%	45%	75%	No	25%	No
Sant Llorenç	5600-5200	G & P	Cave	1	100%	0	0	0%	0%	0%	No	No	No	No
El Cavet	5600-5200	S & E	Open air	22	68%	27%	5%	27%	14%	63%	86%	No	14%	No
La Serreta	5500	G & P	Open air	12	58%	42%	0	50%	66%	91%	71%	29%	No	No
Can Sadurní	5500	G & P	Cave	3	66%	33%	0	0	33%	100%	25%*	25%*	50%*	Yes
Font Major	5400-5000	S & E	Cave	5	100%	0	0	40%	0	100%	No	100%	No	No
Guineu	5200-5000	G & P	Cave	13	62%	38%	0	69%	62%	92%	71%	29%	No	No
Vidre	5400-5000	S & E	Cave	6	50%	50%	0	33%	33%	100%	No	No	100%	Yes
Cau Guilles	Cardial	Cap de Creus	Cave	4	0	100%	0	25%	75%	50%	100%	No	No	No
EPICARDIAL				76										
Barranc Fabra	5027-4488	S & E	Open air	10	40%	50%	10%	10%	10%	80%	100%	No	No	No
Molló	Epicardial	S & E	Open air	7	43%	57%	0	29%	29%	100%	100%	No	No	No
Can Sadurní	Epicardial	G & P	Cave	4	75%	25%	0	75%	25%	100%	10%*	No*	90%*	Yes
CRA	4700-4500	Barcelona	Open air	52	96%	3%	3%	57.6%	0%	77%	80%	No	20%	Yes
Sant Llorenç	5200-5400	G & P	Cave	3	33%	33%	33%	66%	0%	100%	100%	No	No	No
BEYOND				31										
Can Sadurní	4500-2000	G & P	Cave	14	86%	14%	0	42%	0	100%	-	-	-	Yes

N: Number of vessels analysed

Residues: Percentage of samples with significant residues.

>250°C: Sites with samples containing biomarkers for temperatures higher than 250°C.

*: Results incorporating data from Spiteri et al 2016.

Regions: G & P: Garraf massif and Penedès valley. S & E: South and Ebro valley.

Appendix 6

Cromatogram abbreviations:

C34, C36 (IS): internal standards

Dx (where x is a number): dicarboxylic acids with x carbons in the chain

P: phthalate plasticiser

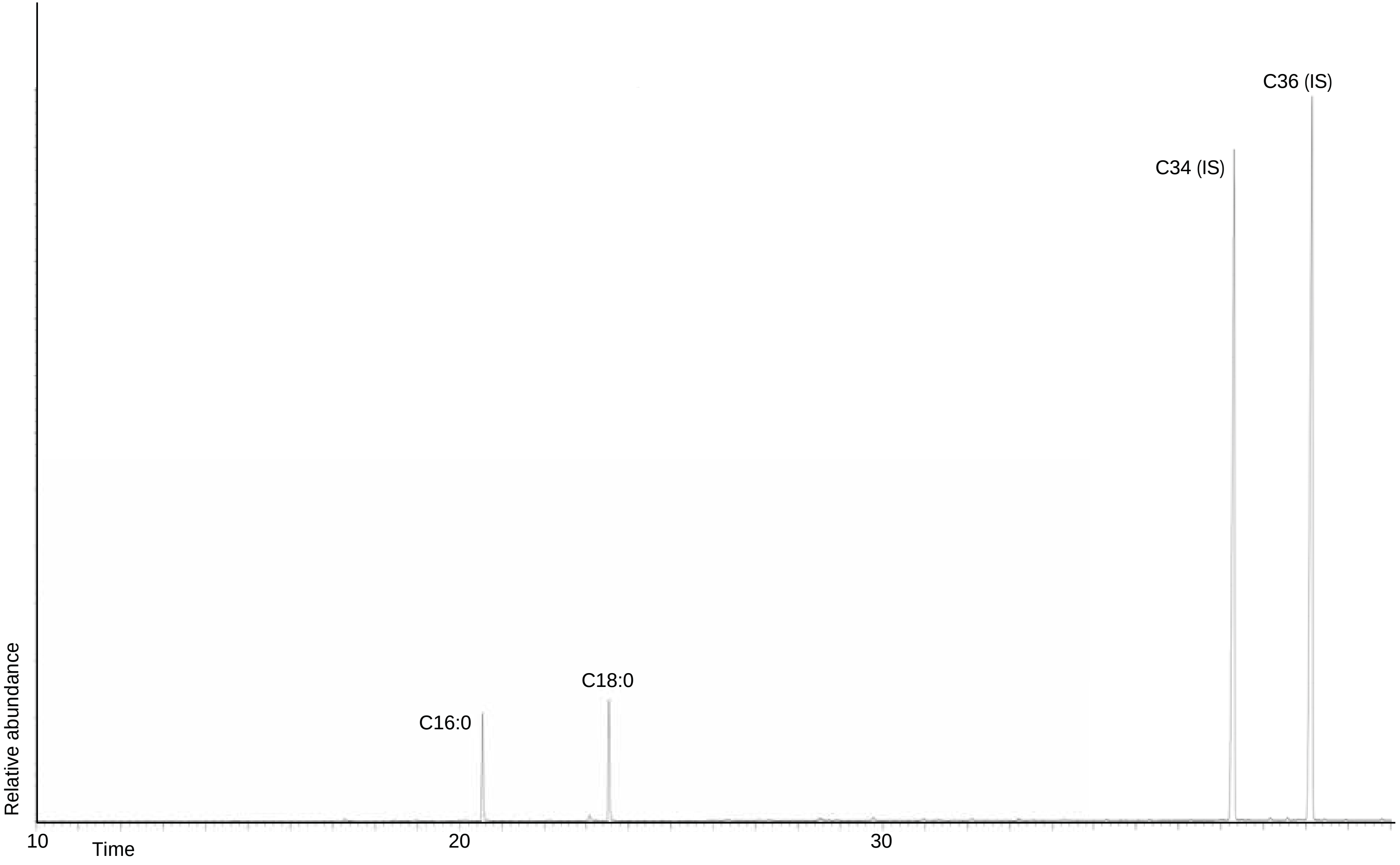
▪ : Alkane

CFAM: Cyclopentyl fatty acid

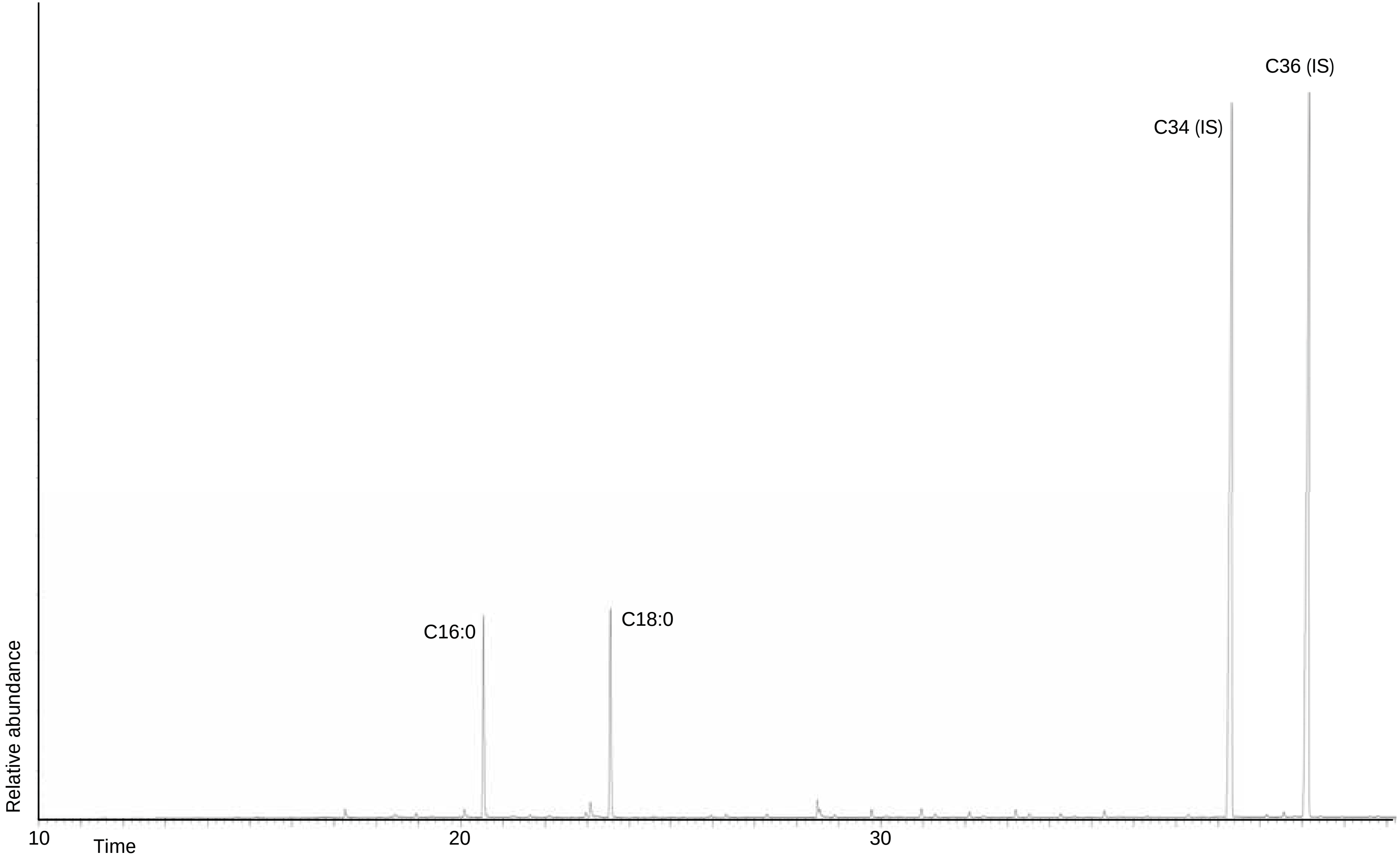
Ax (where x is a number): fatty alcohol with x carbons in the chain

Berenguer de Palou

BDP 438

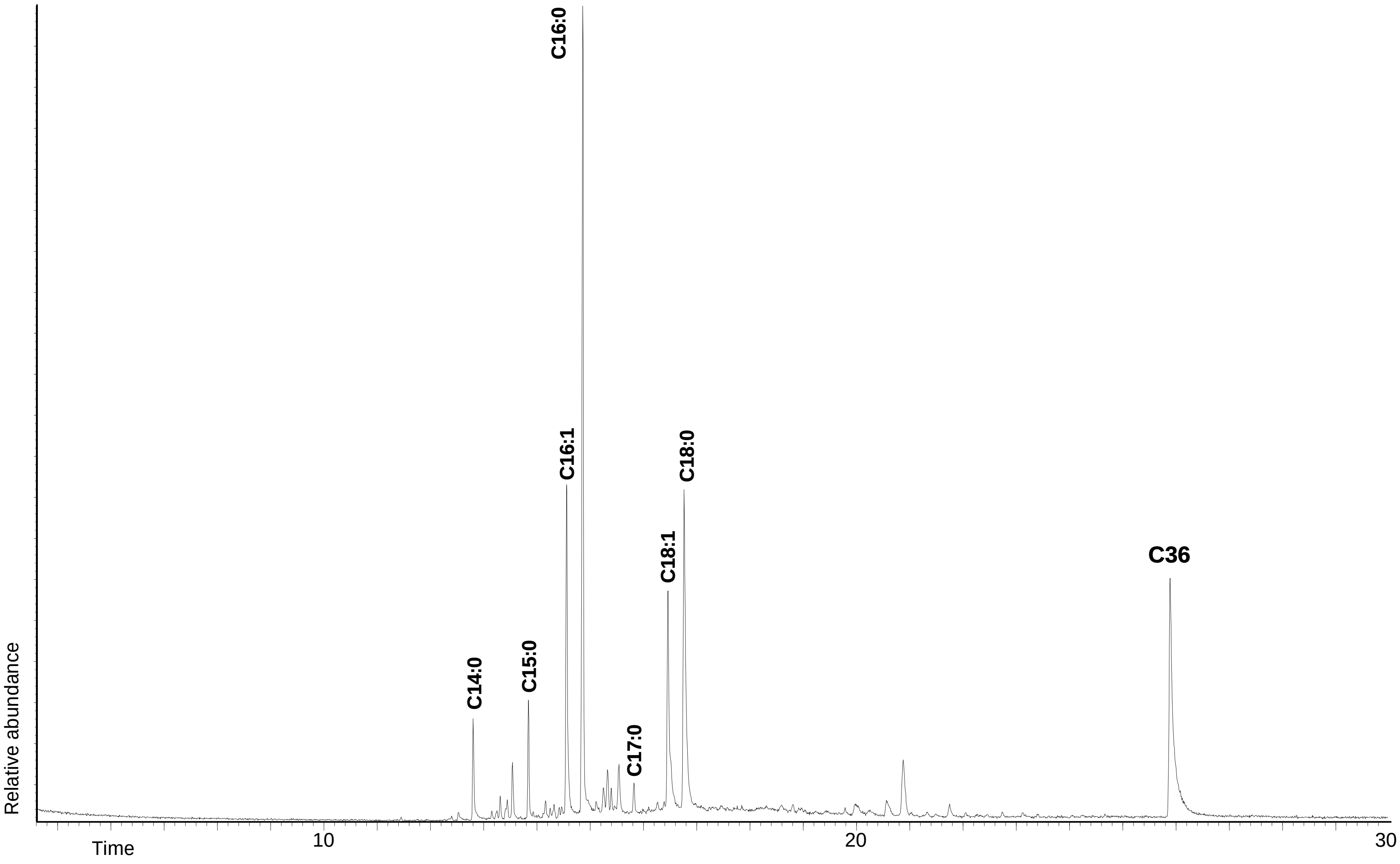


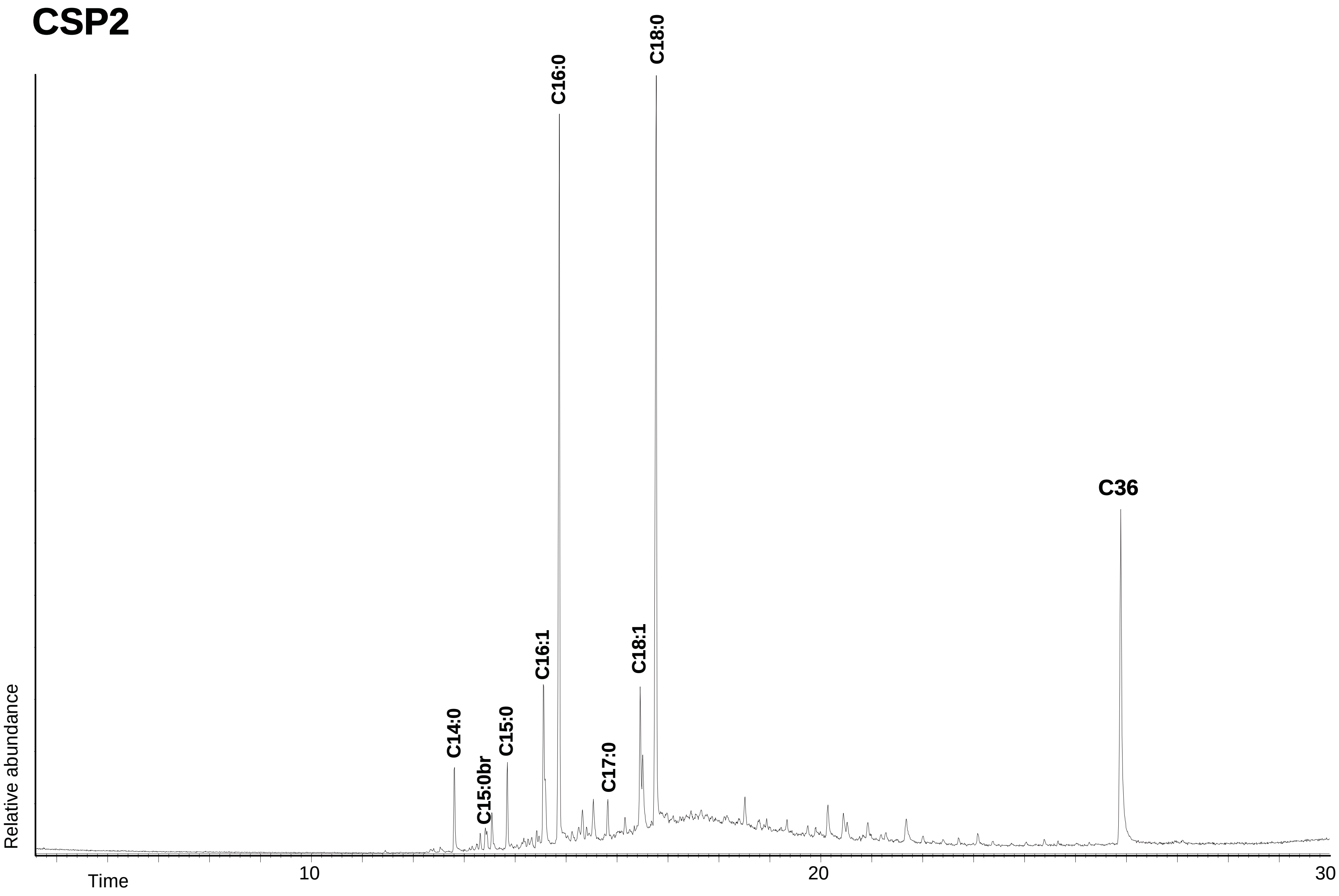
BDP 458

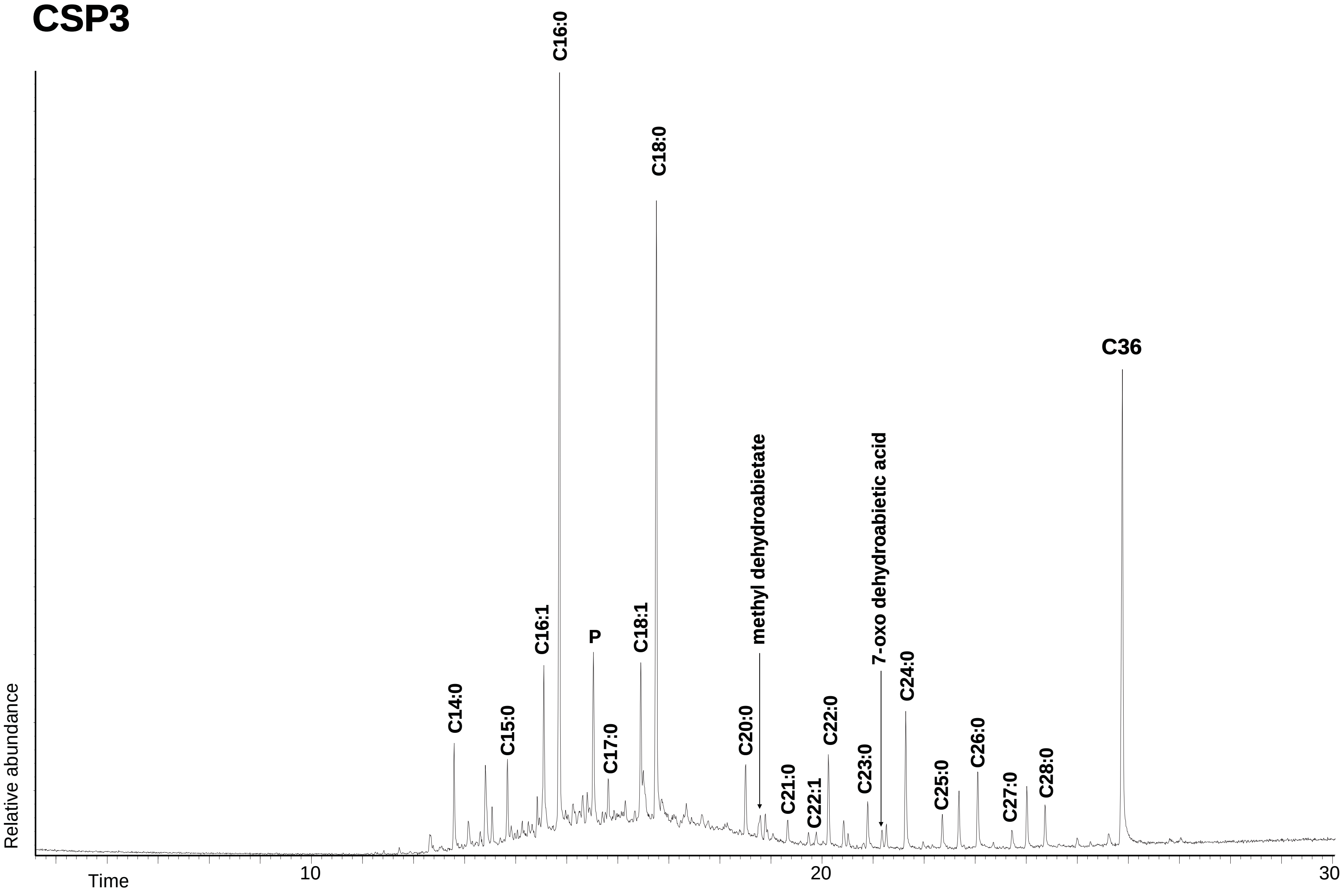


Caserna de Sant Pau

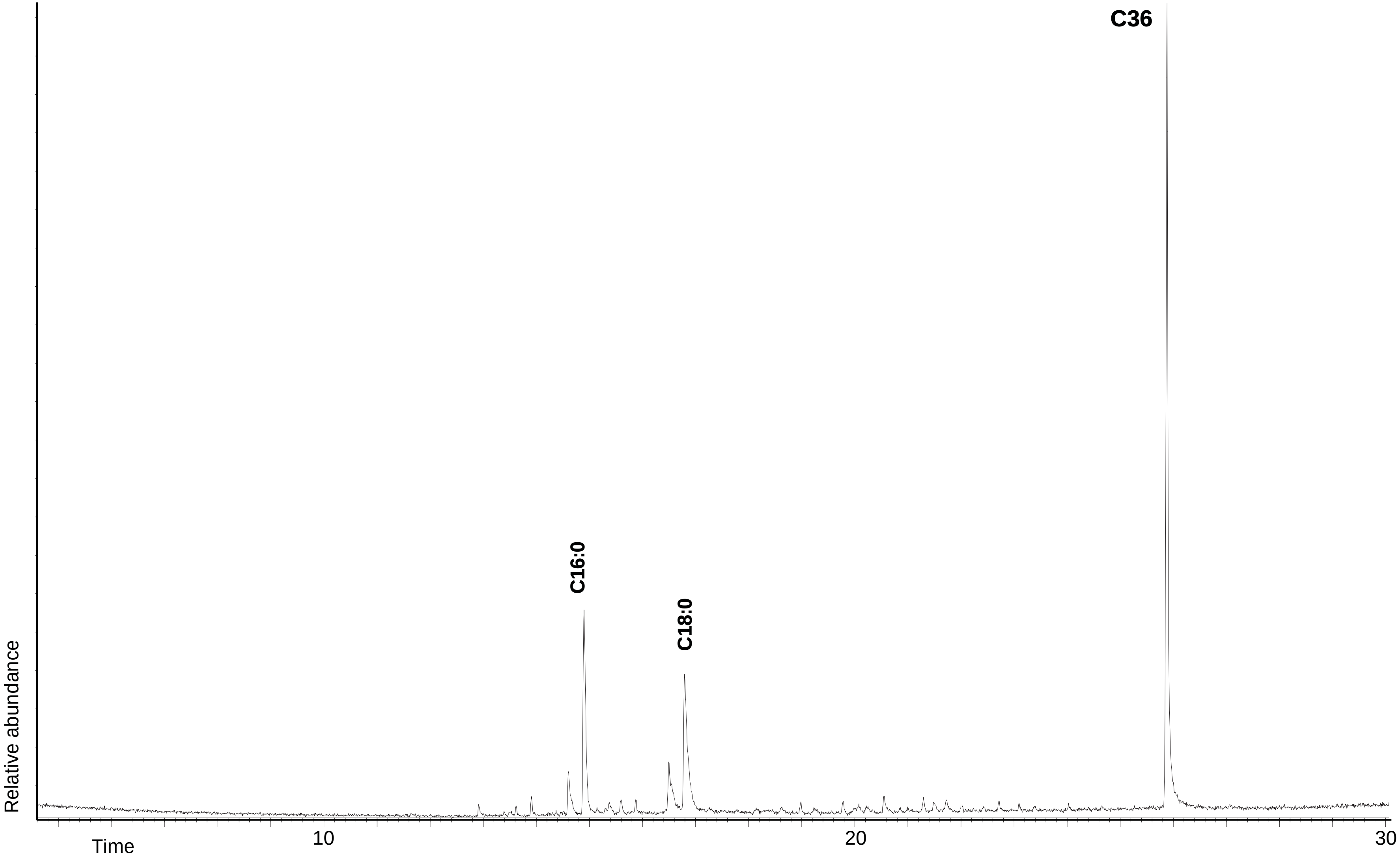
CSP1


C16:0
C16:1
C18:0
C18:1
C36
C14:0
C15:0
C17:0
Relative abundance
Time
10
20
30

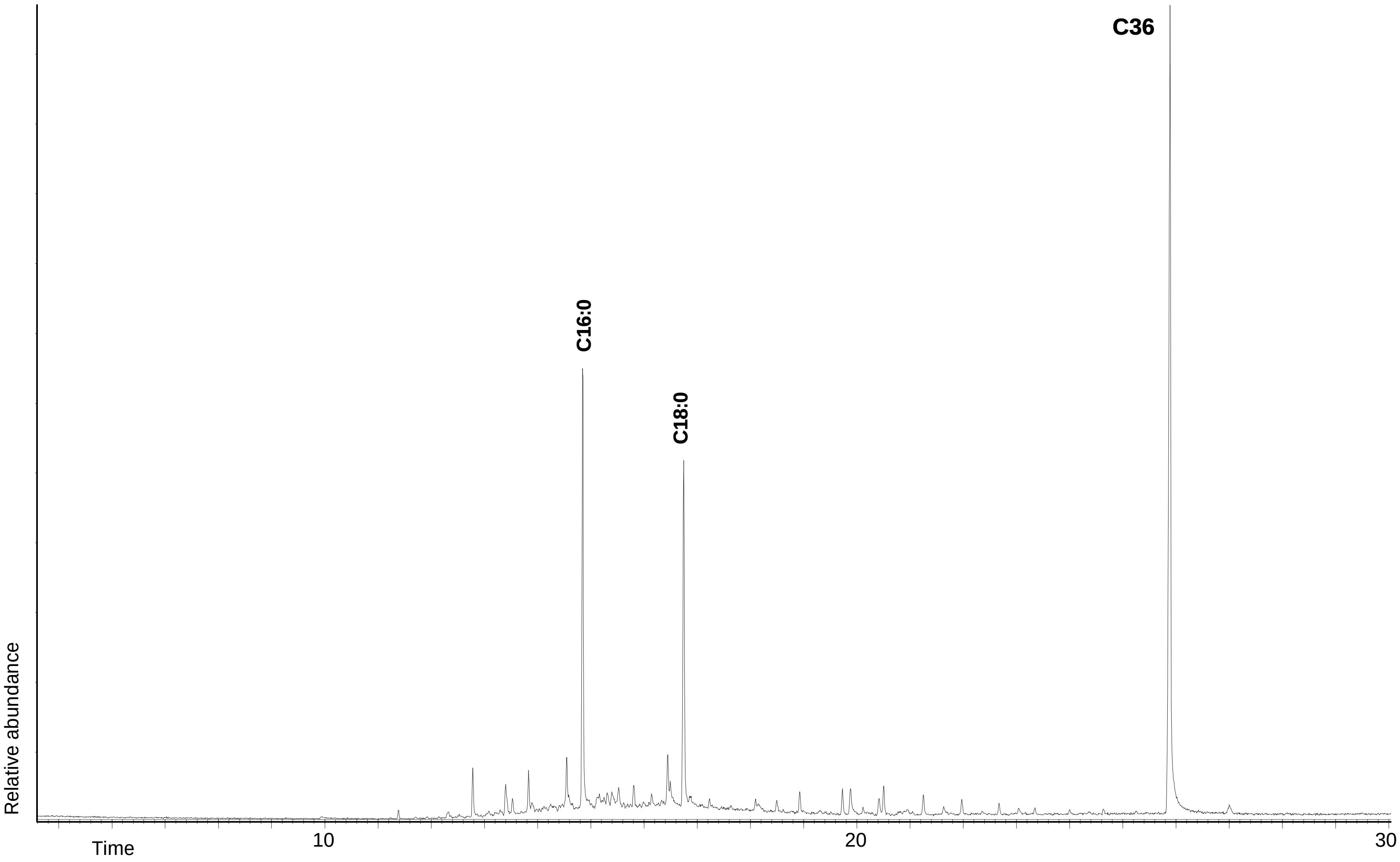

CSP2
C18:0
C16:0
C36
C16:1
C18:1
C14:0
C15:0
C17:0
C15:0br
Relative abundance
Time
10
20
30


CSP3
C16:0
C18:0
C36
methyl dehydroabietate
7-oxo dehydroabietic acid
C16:1
P
C18:1
C24:0
C14:0
C15:0
C17:0
C20:0
C22:0
C26:0
C23:0
C21:0
C25:0
C22:1
C27:0
C28:0
Relative abundance
Time
10
20
30

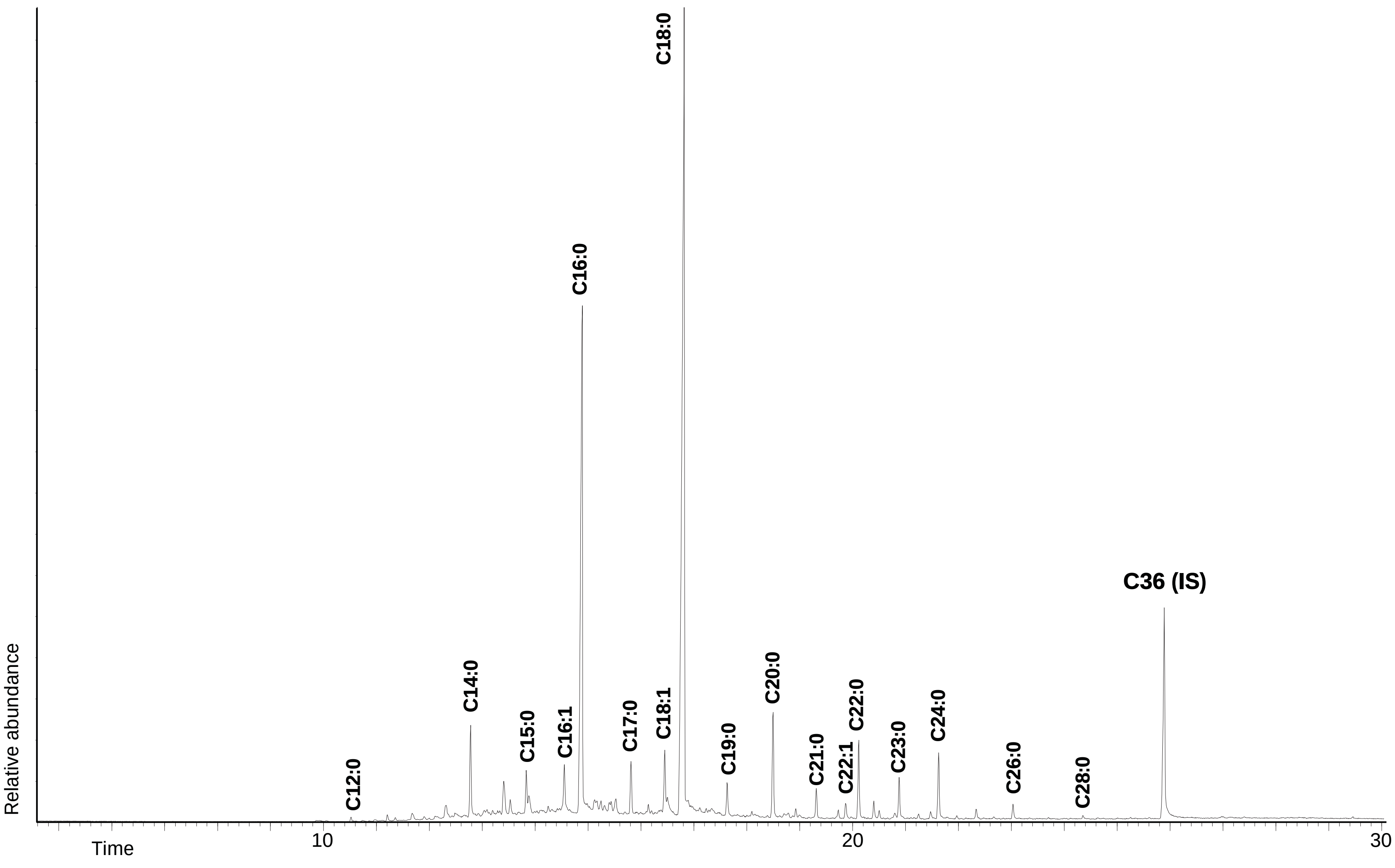
CSP4



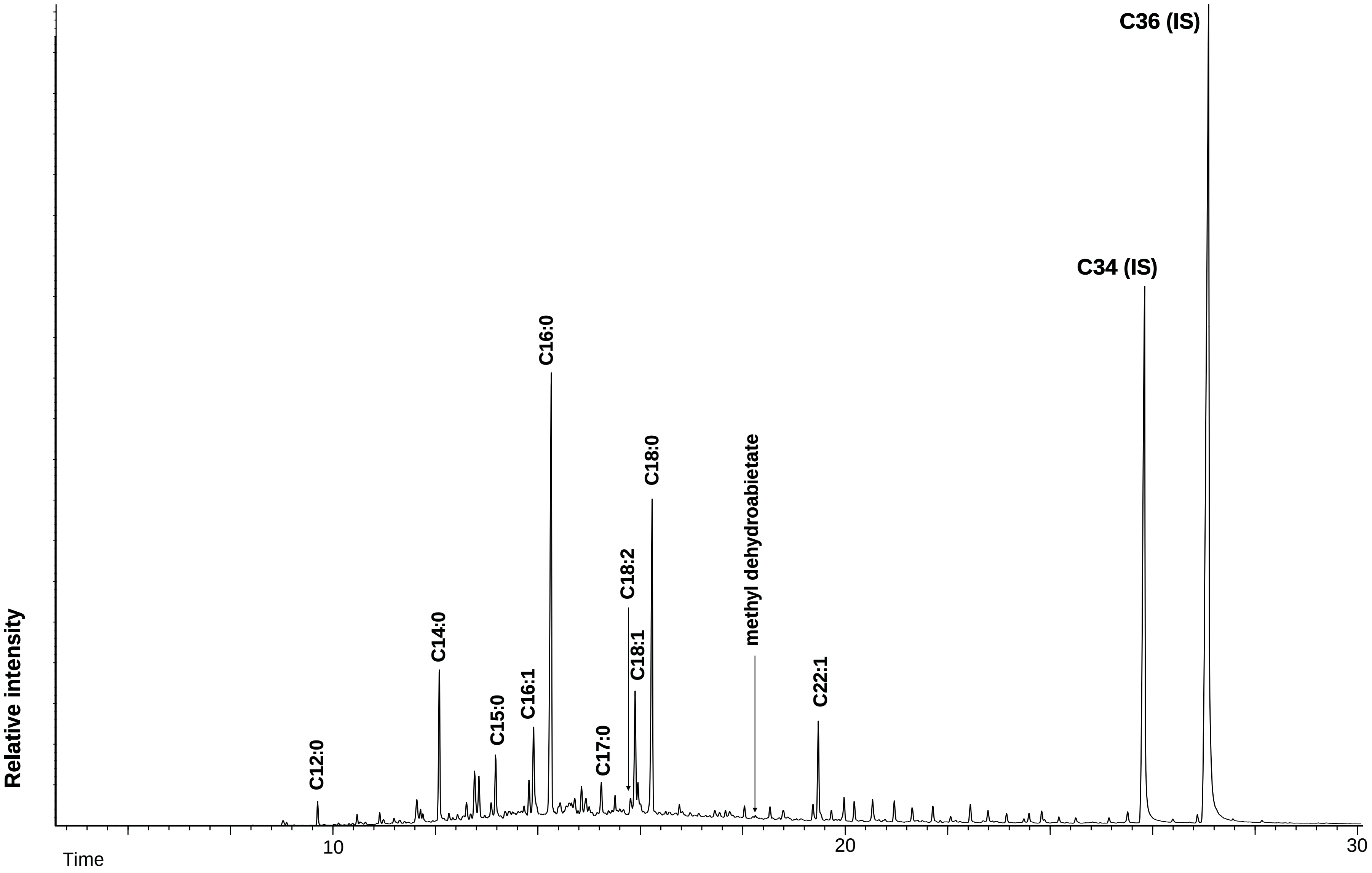
CSP5



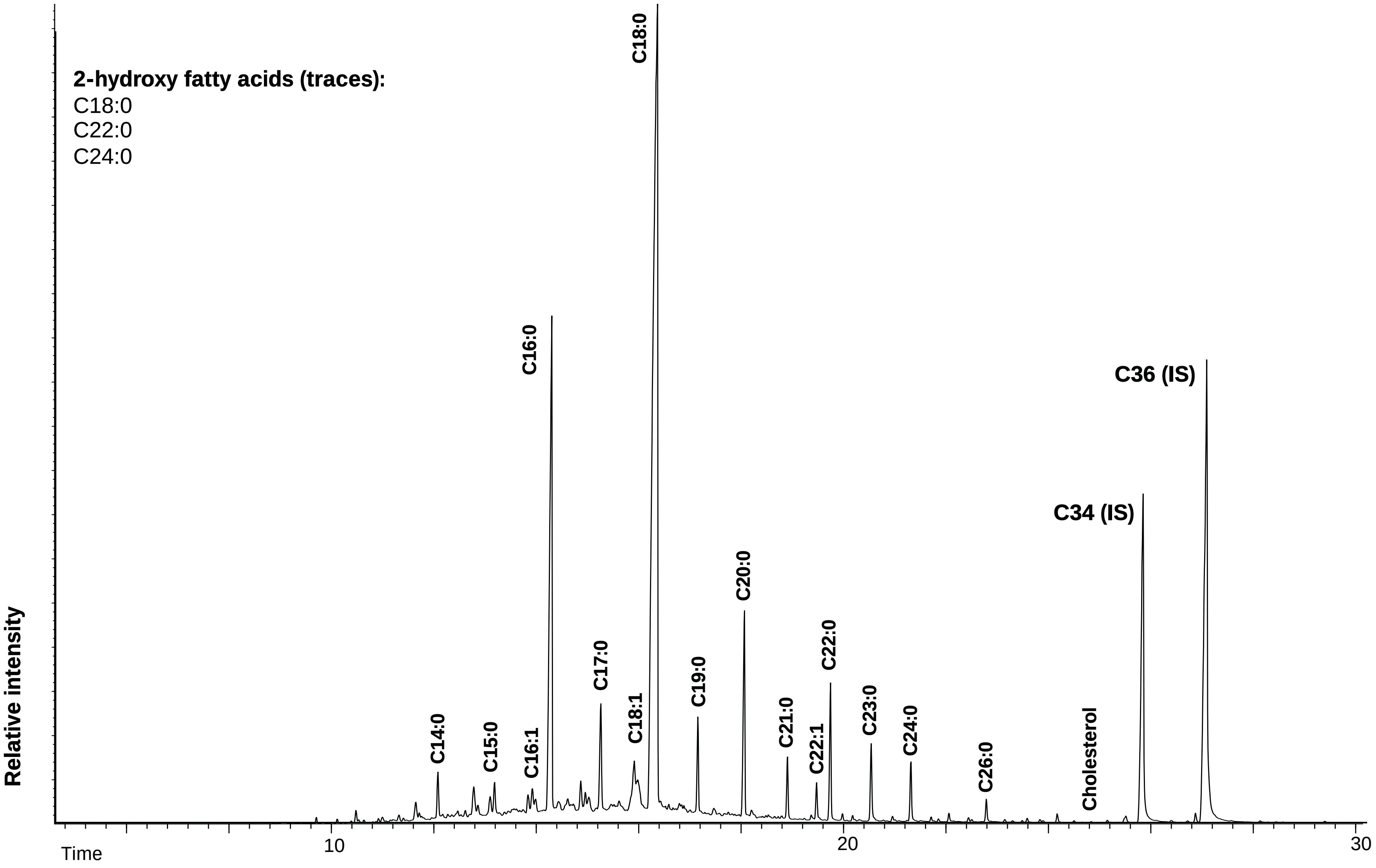
CSP6


C18:0
C16:0
C36 (IS)
C12:0
C14:0
C15:0
C16:1
C17:0
C18:1
C19:0
C20:0
C21:0
C22:1
C22:0
C23:0
C24:0
C26:0
C28:0
Relative abundance
Time
10
20
30

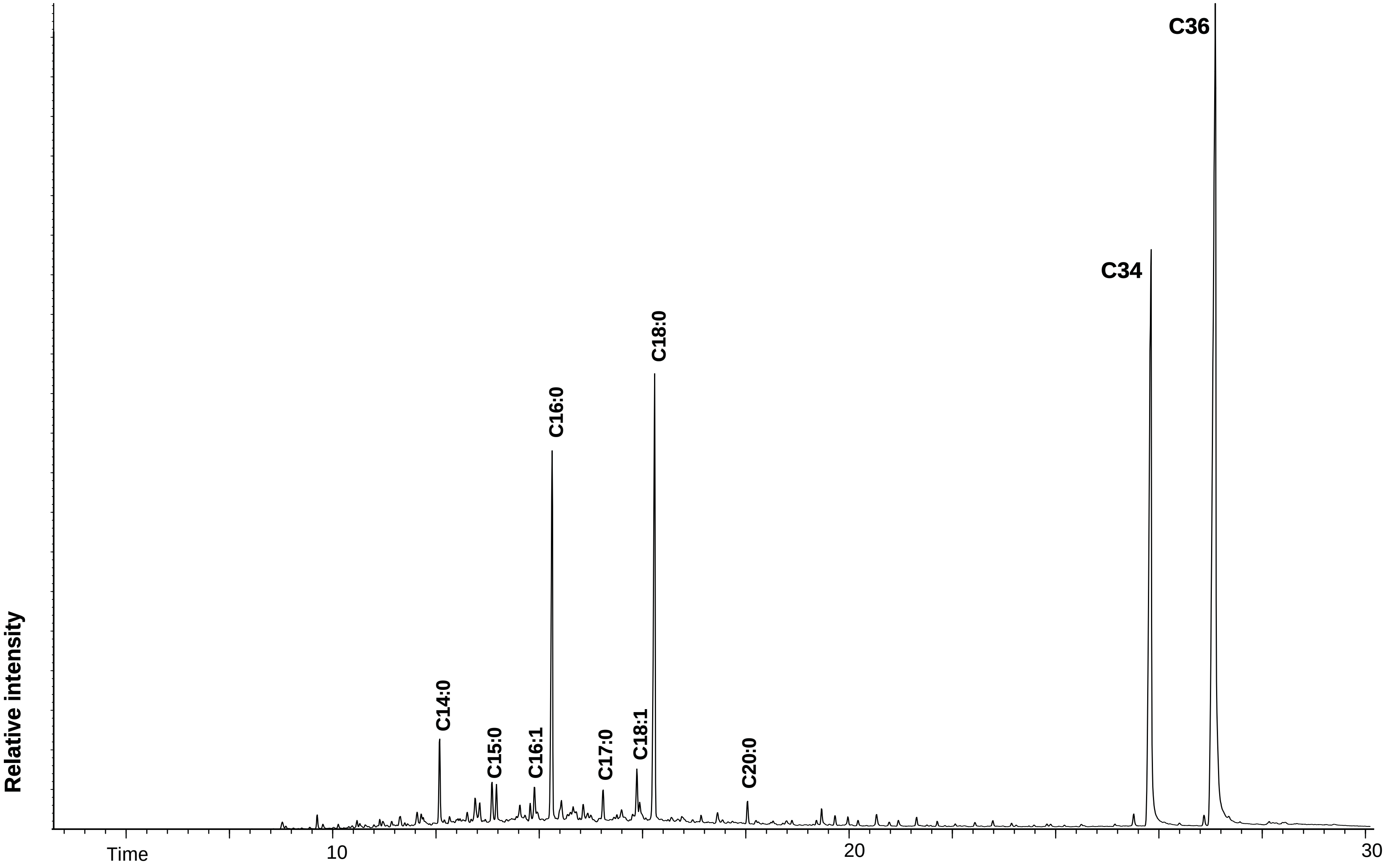
CSP7


C36 (IS)
C34 (IS)
C16:0
C18:0
methyl dehydroabietate
C18:2
C14:0
C18:1
C22:1
C16:1
C15:0
C17:0
C12:0
Relative intensity
Time
10
20
30

CSP8

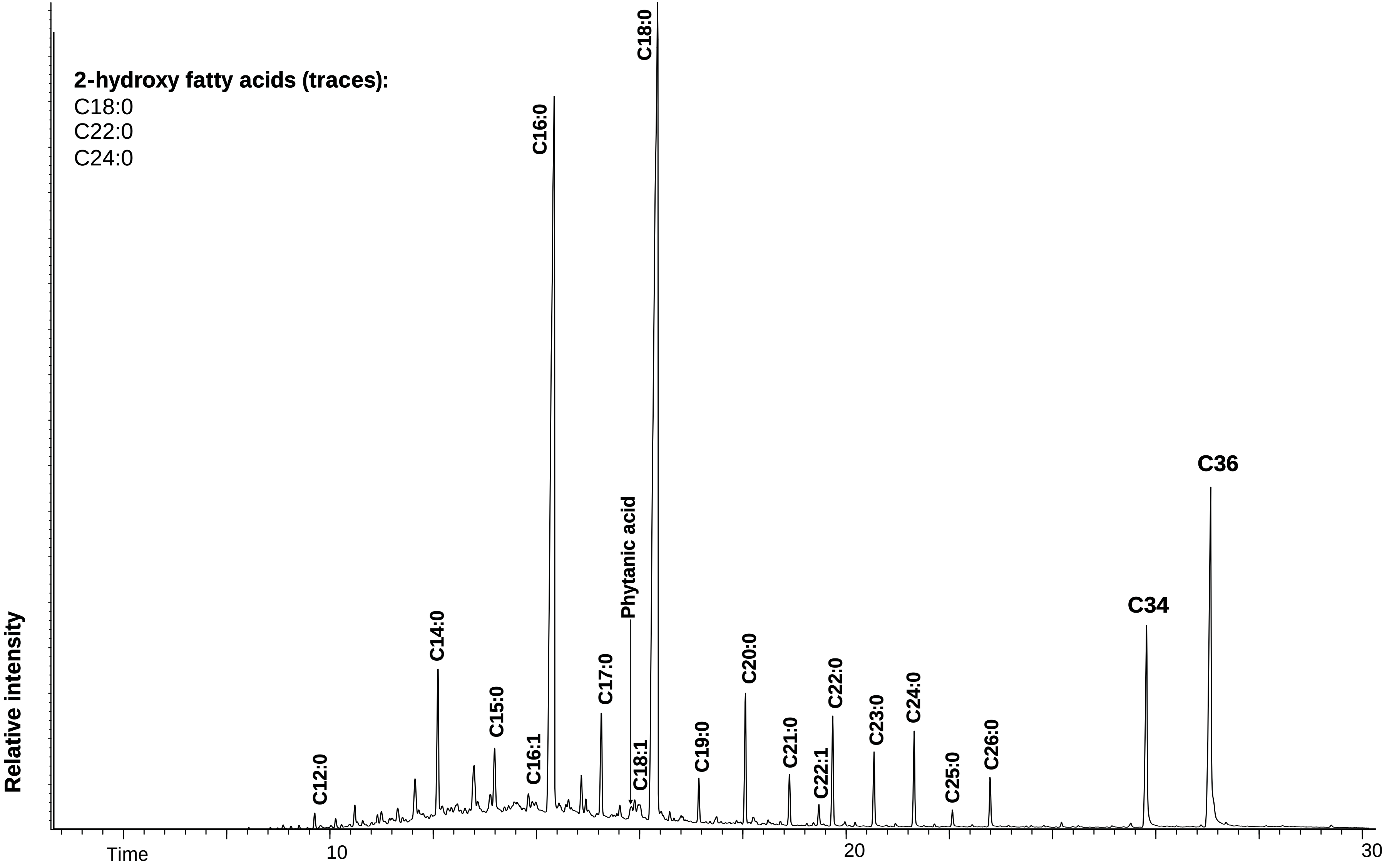

2-hydroxy fatty acids (traces):
C18:0
C22:0
C24:0
Relative intensity
Time
10
20
30
C14:0
C15:0
C16:1
C16:0
C17:0
C18:1
C18:0
C19:0
C20:0
C21:0
C22:1
C22:0
C23:0
C24:0
C26:0
Cholesterol
C34 (IS)
C36 (IS)

CSP9

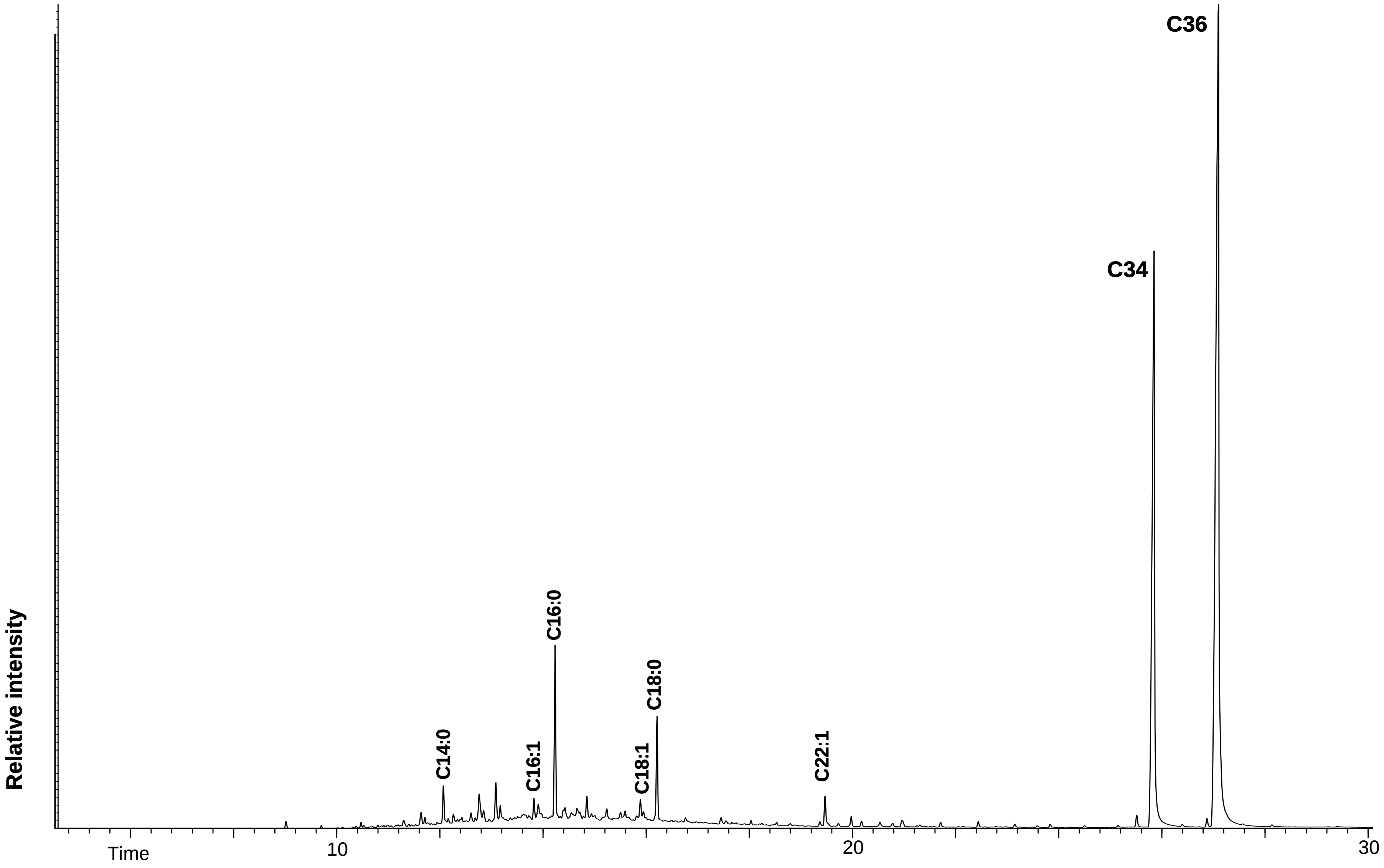


Relative intensity
Time
10
20
30
C14:0
C15:0
C16:1
C16:0
C17:0
C18:1
C18:0
C20:0
C34
C36

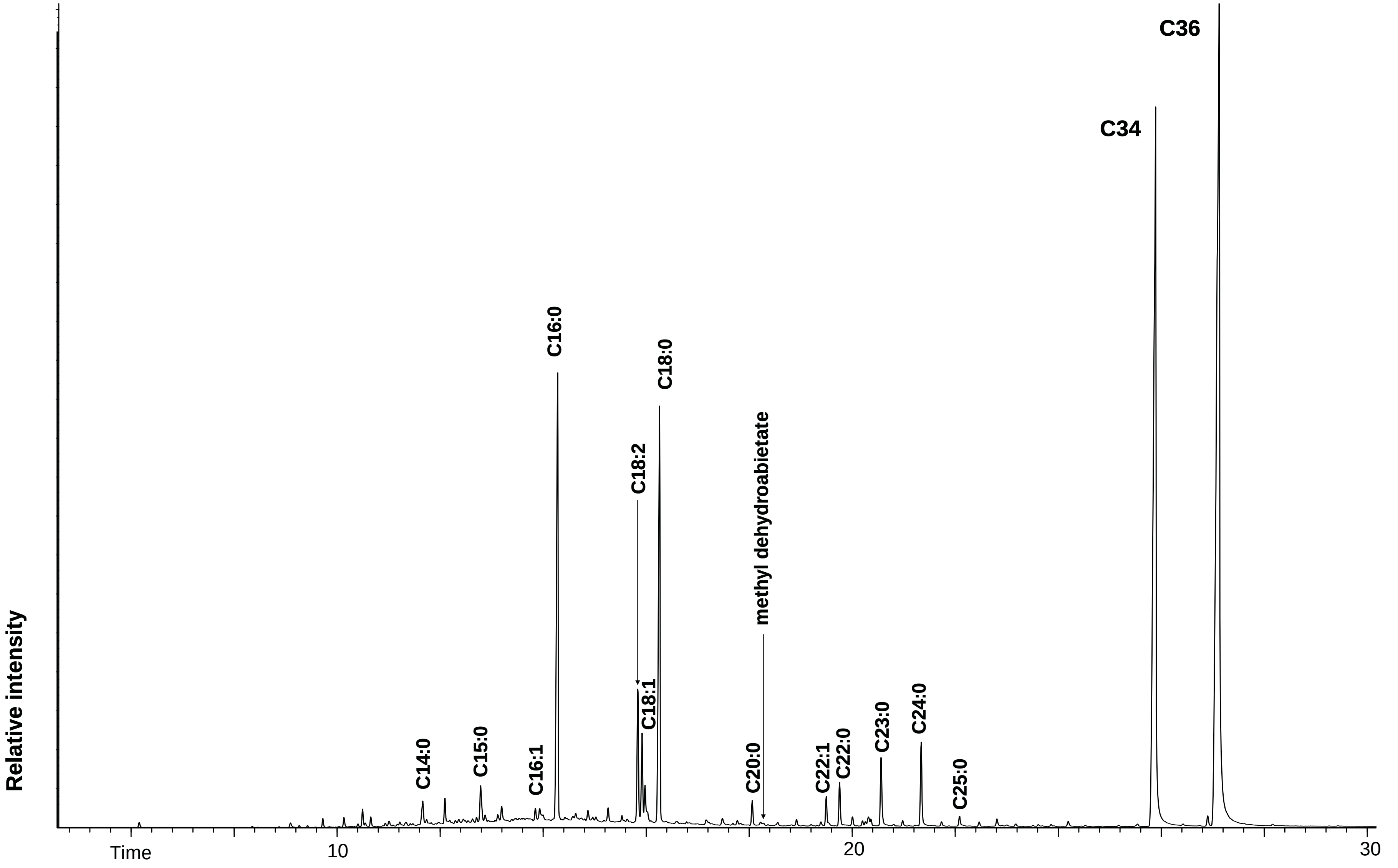
CSP10

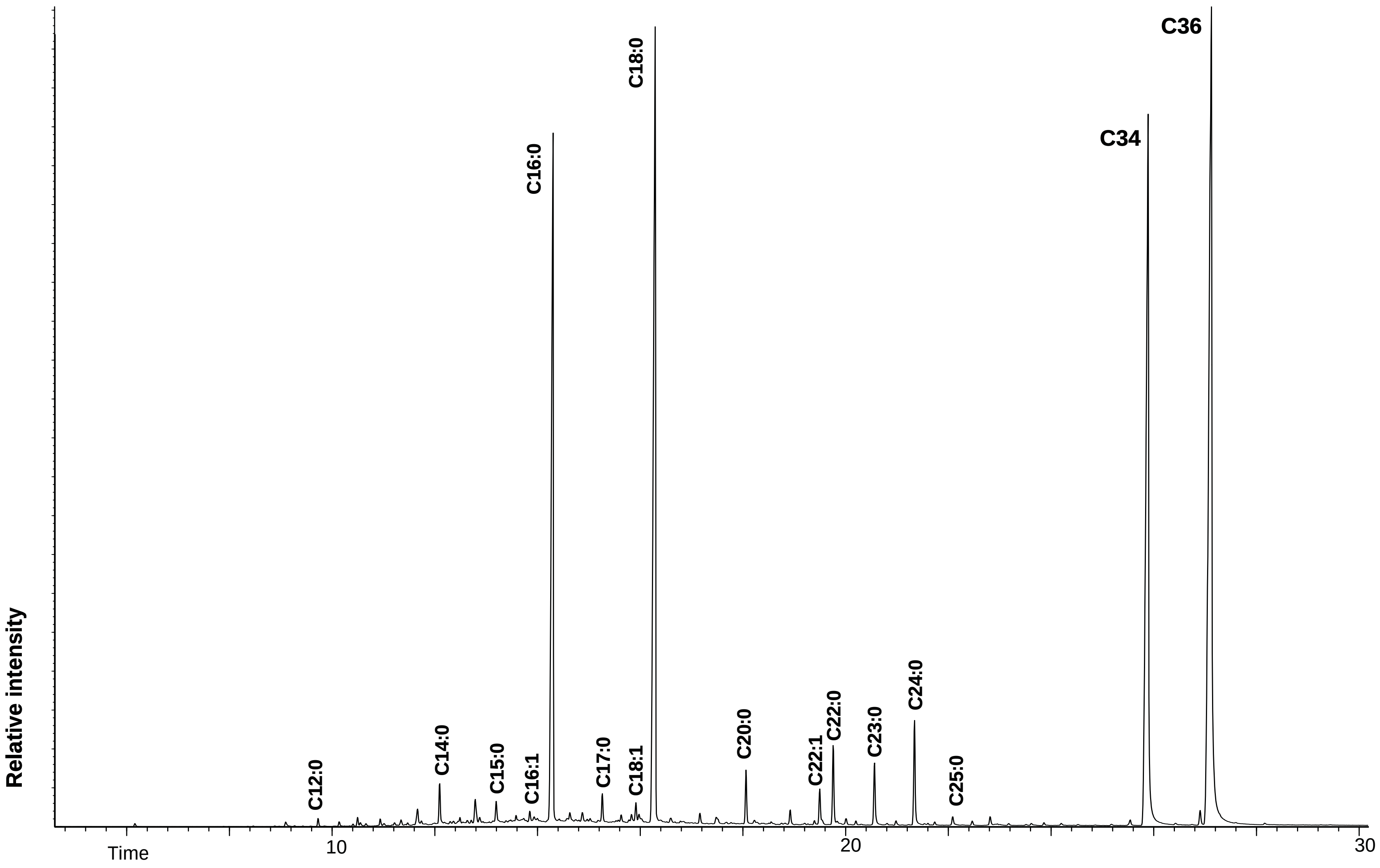

2-hydroxy fatty acids (traces):
C18:0
C22:0
C24:0
Relative intensity
Time
10
20
30
C12:0
C14:0
C15:0
C16:1
C16:0
C17:0
Phytanic acid
C18:1
C18:0
C19:0
C20:0
C21:0
C22:1
C22:0
C23:0
C24:0
C25:0
C26:0
C34
C36

CSP11


C36
C34
C16:0
C18:0
C14:0
C16:1
C18:1
C22:1
Relative intensity
Time
10
20
30

CSP12

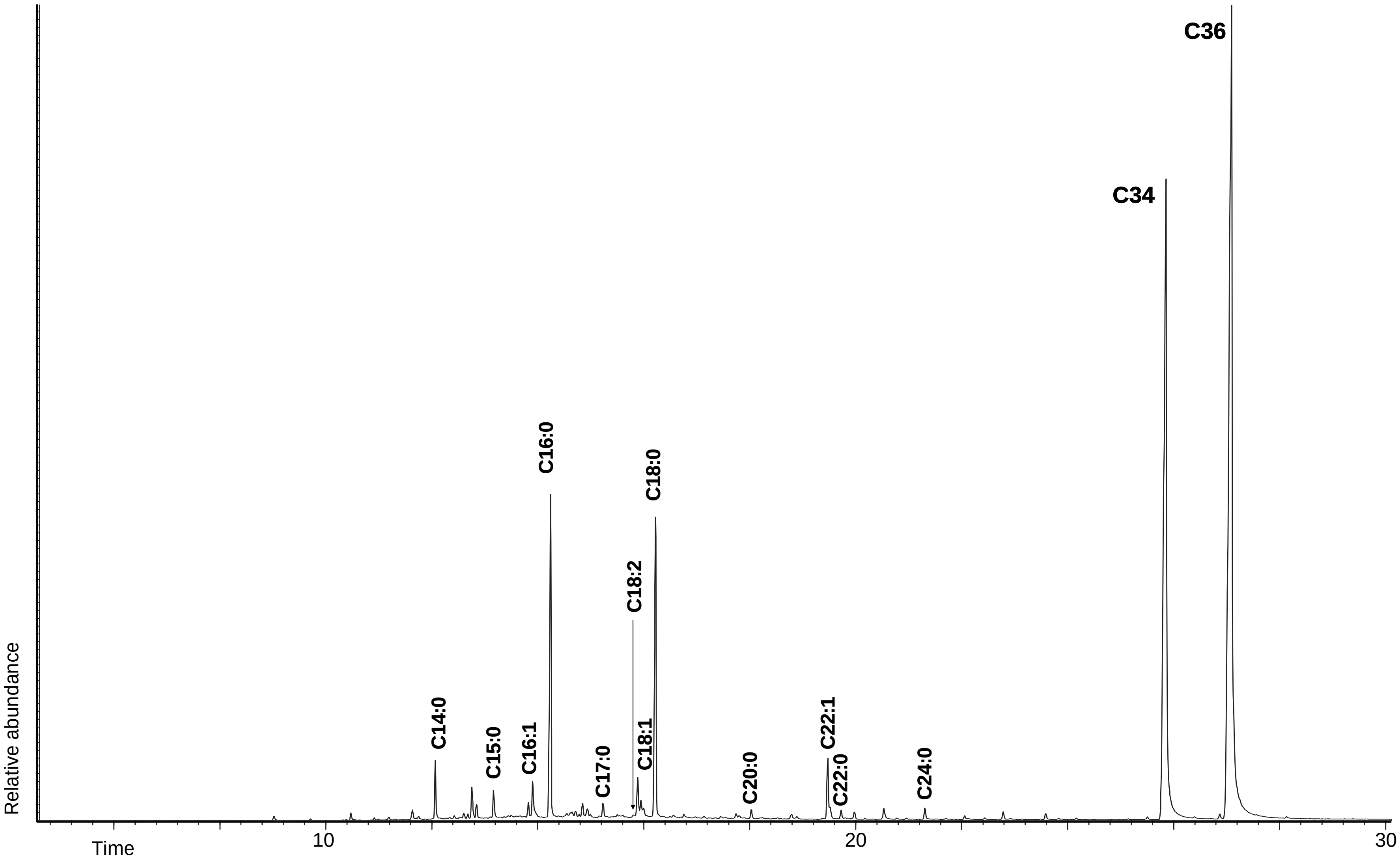

C36
C34
C16:0
C18:0
C18:2
methyl dehydroabietate
C18:1
C14:0
C15:0
C16:1
C20:0
C22:1
C22:0
C23:0
C24:0
C25:0
Relative intensity
Time
10
20
30

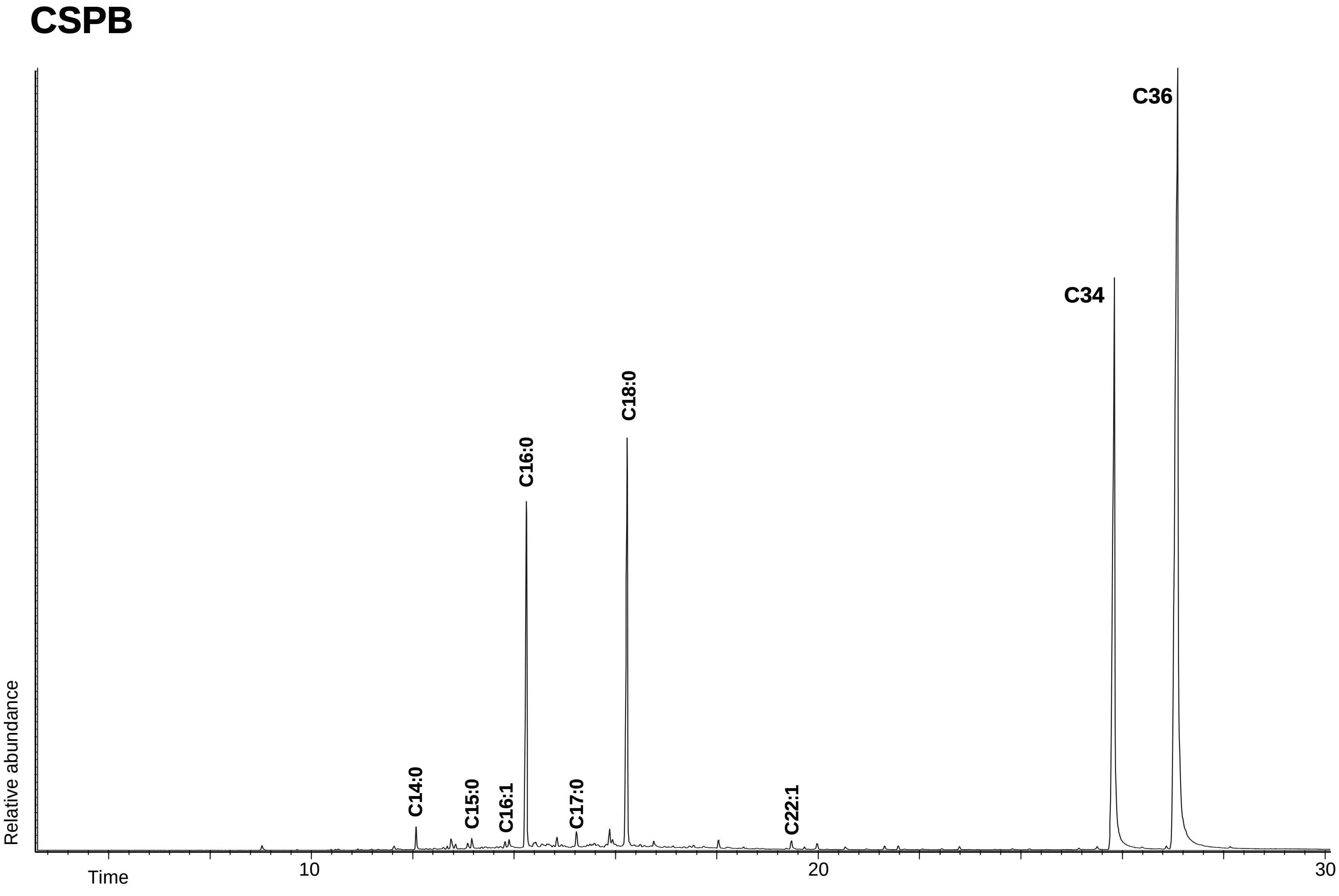


CSP13

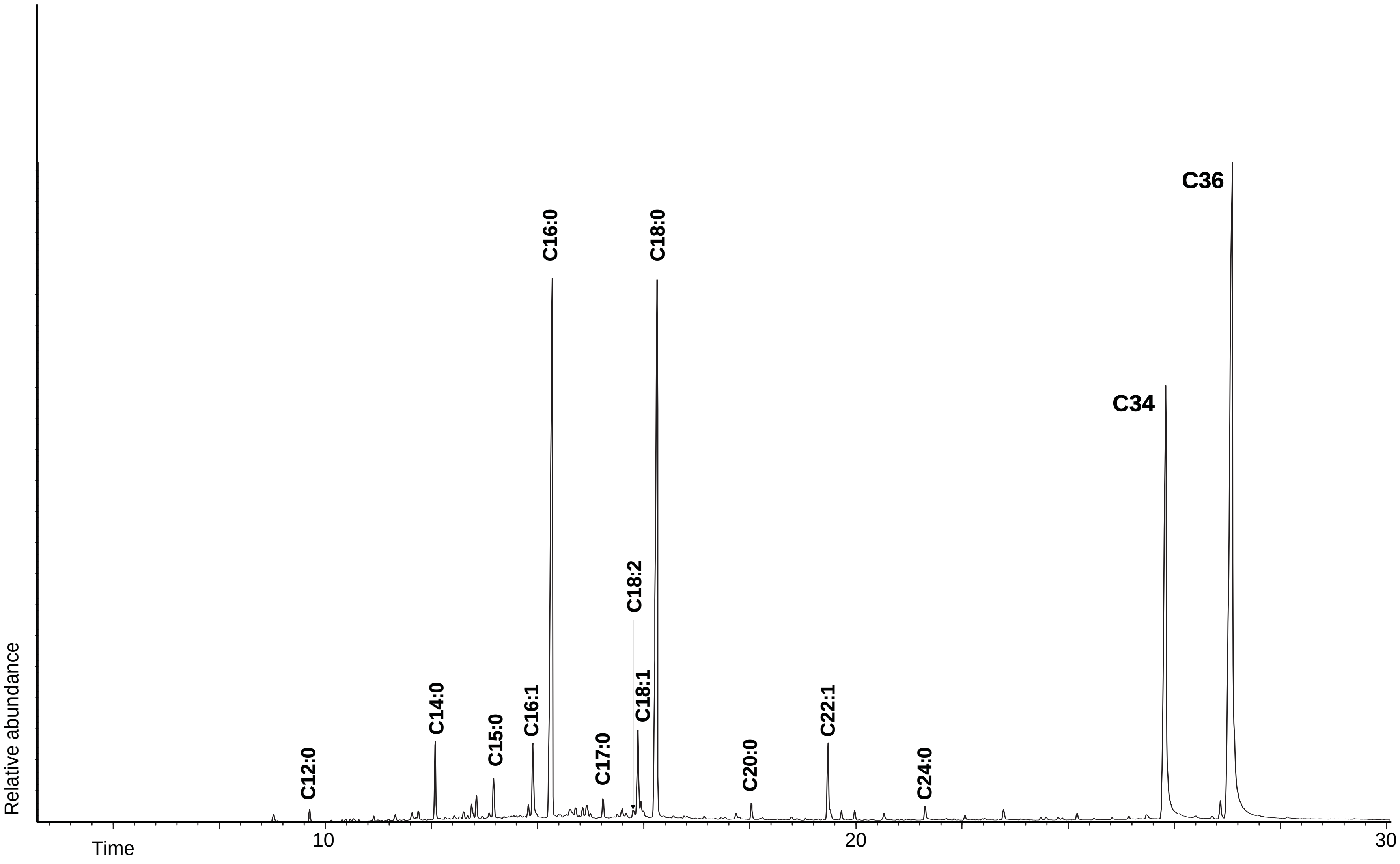
C36
C18:0
C34
C16:0
C12:0
C14:0
C15:0
C16:1
C17:0
C18:1
C20:0
C22:1
C22:0
C23:0
C24:0
C25:0
Relative intensity
Time
10
20
30

CSPA

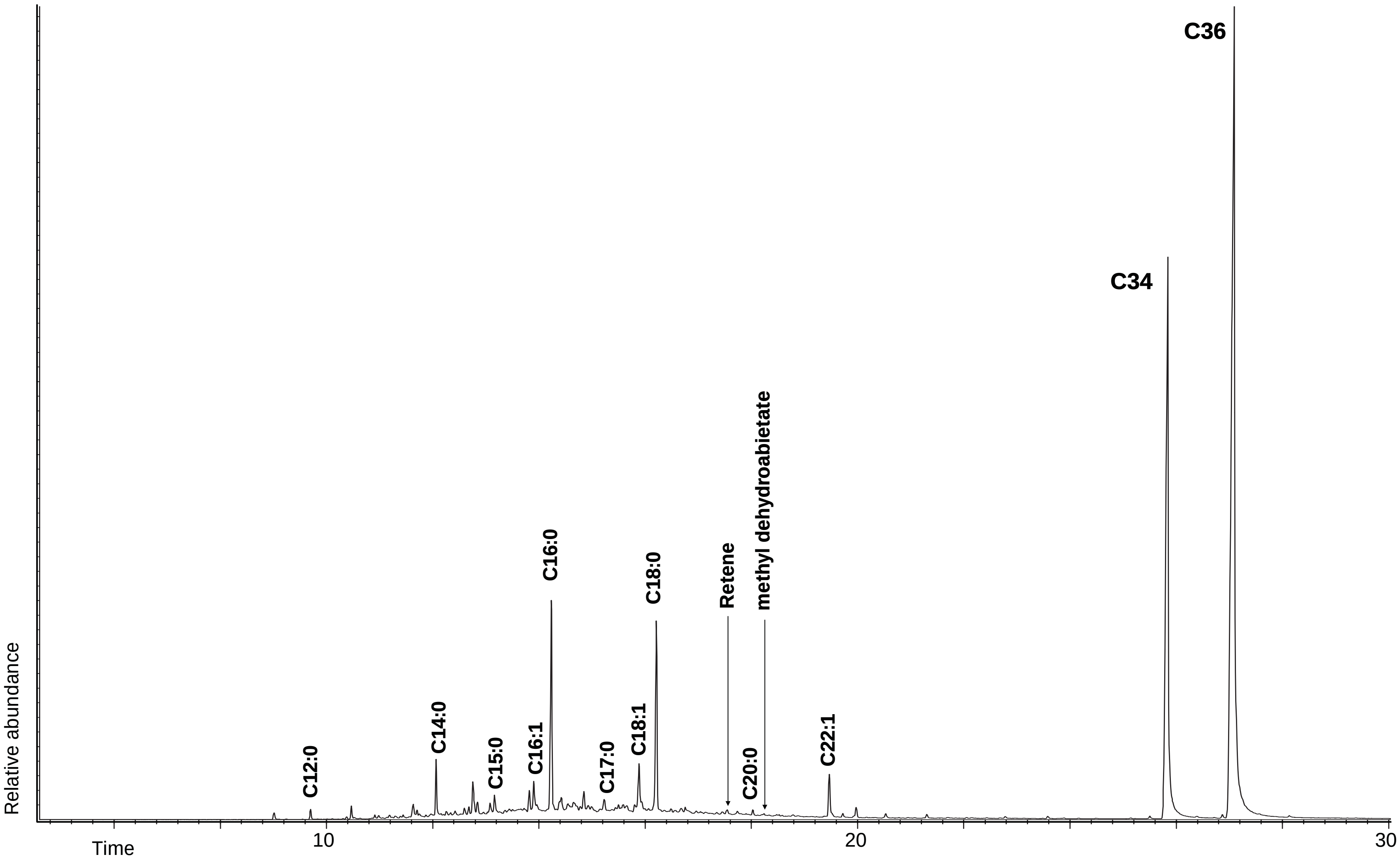

C36
C34
C16:0
C18:0
C18:2
C14:0
C15:0
C16:1
C17:0
C18:1
C20:0
C22:1
C22:0
C24:0
Relative abundance
Time
10
20
30


CSPB
C36
C34
C18:0
C16:0
C14:0
C15:0
C16:1
C17:0
C22:1
Relative abundance
Time
10
20
30

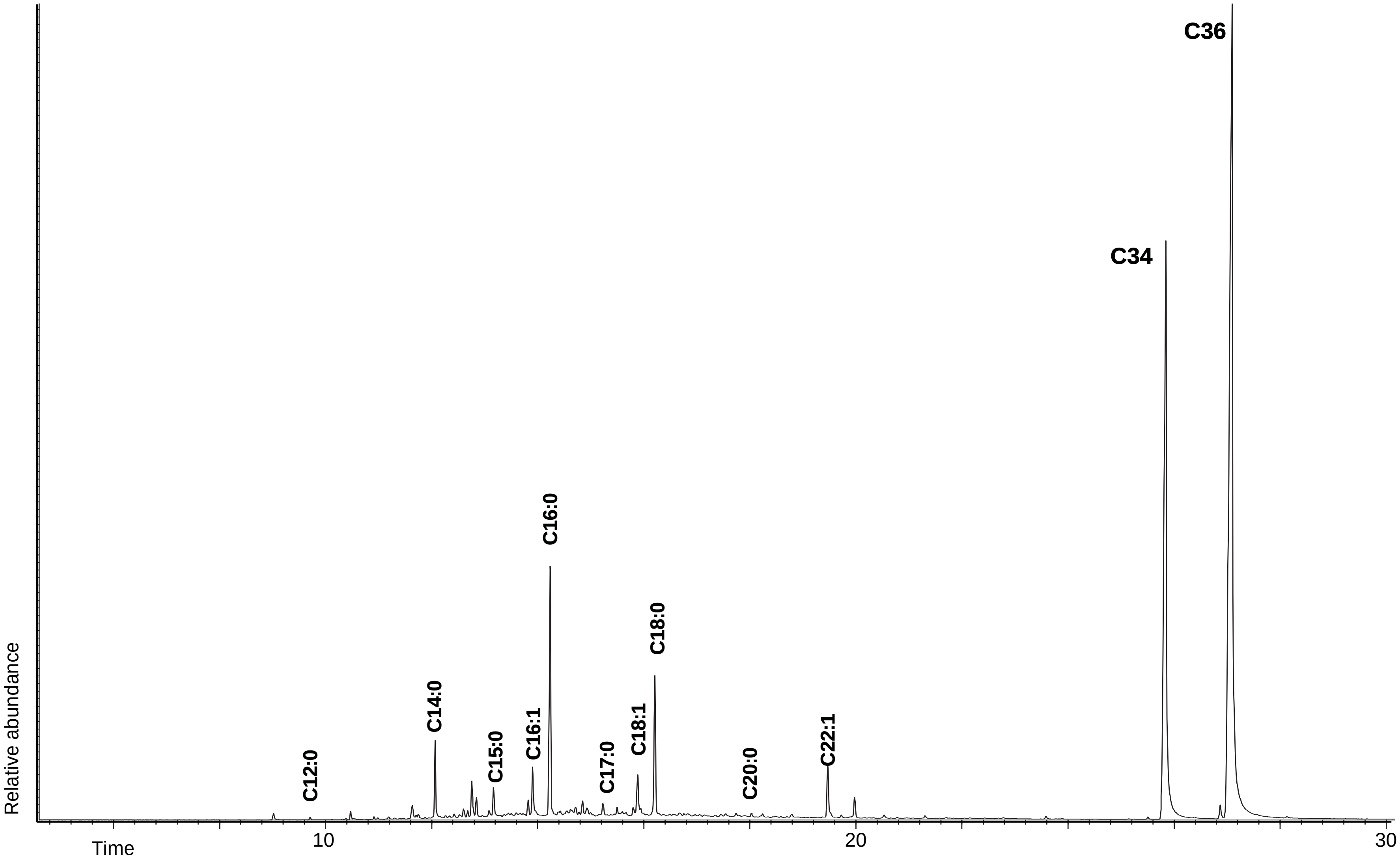
CSPC

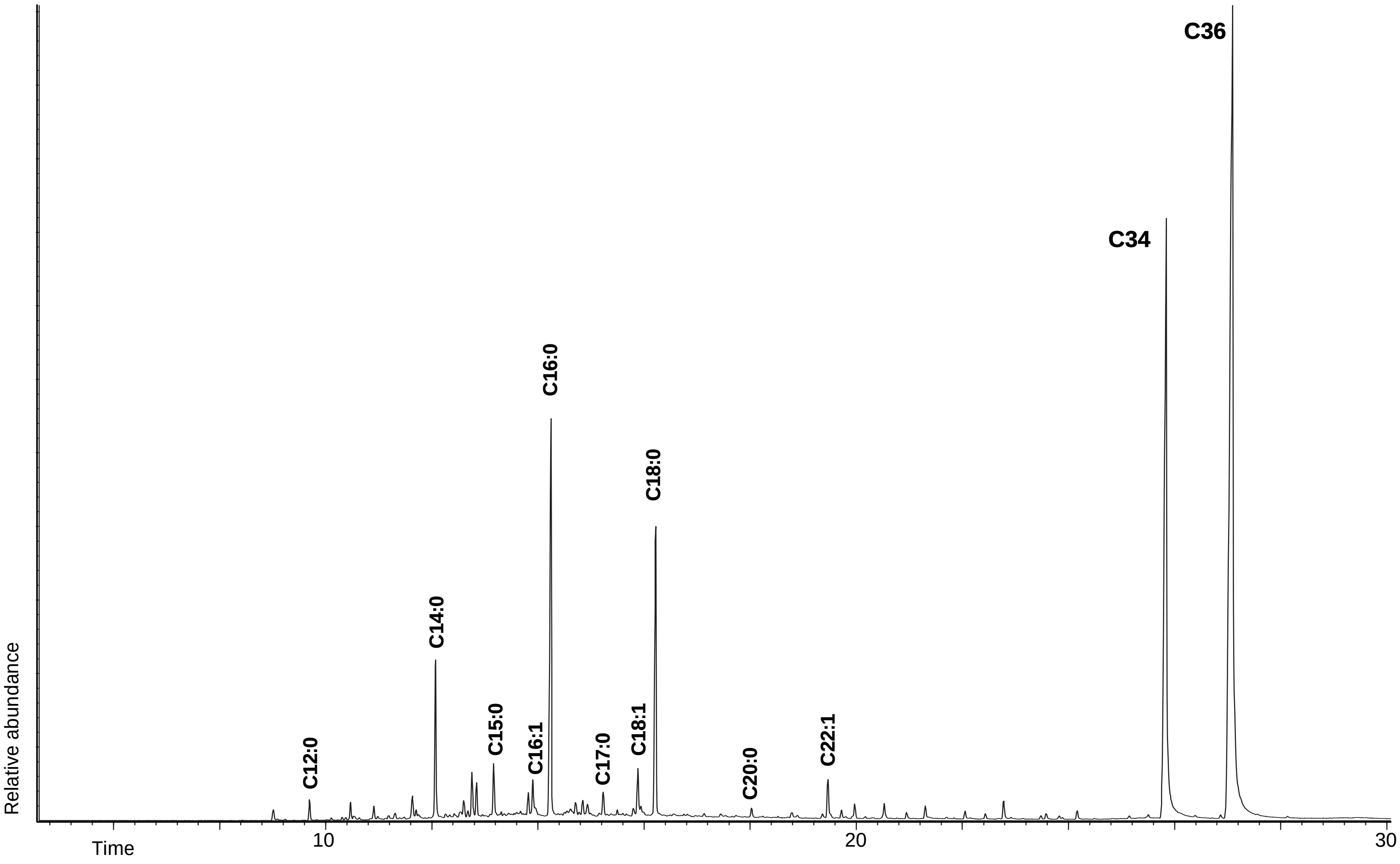

Relative abundance
Time
10
20
30
C12:0
C14:0
C15:0
C16:1
C16:0
C17:0
C18:2
C18:1
C18:0
C20:0
C22:1
C24:0
C34
C36

CSPD


C36
C34
C12:0
C14:0
C15:0
C16:1
C16:0
C17:0
C18:1
C18:0
Retene
C20:0
methyl dehydroabietate
C22:1
Relative abundance
Time
10
20
30

CSPE

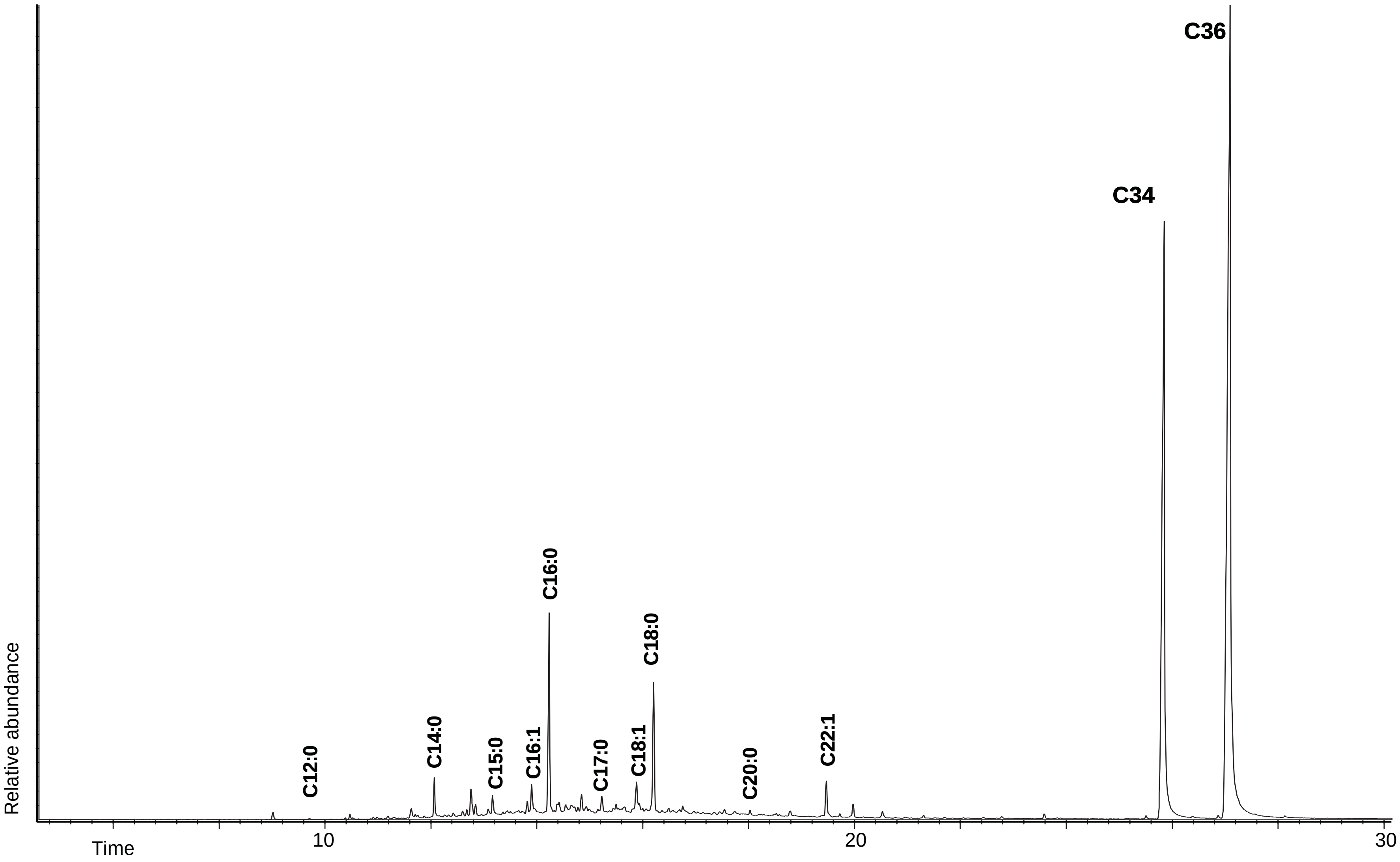

C36
C34
C16:0
C18:0
C14:0
C18:1
C16:1
C22:1
C15:0
C17:0
C20:0
C12:0
Relative abundance
Time
10
20
30



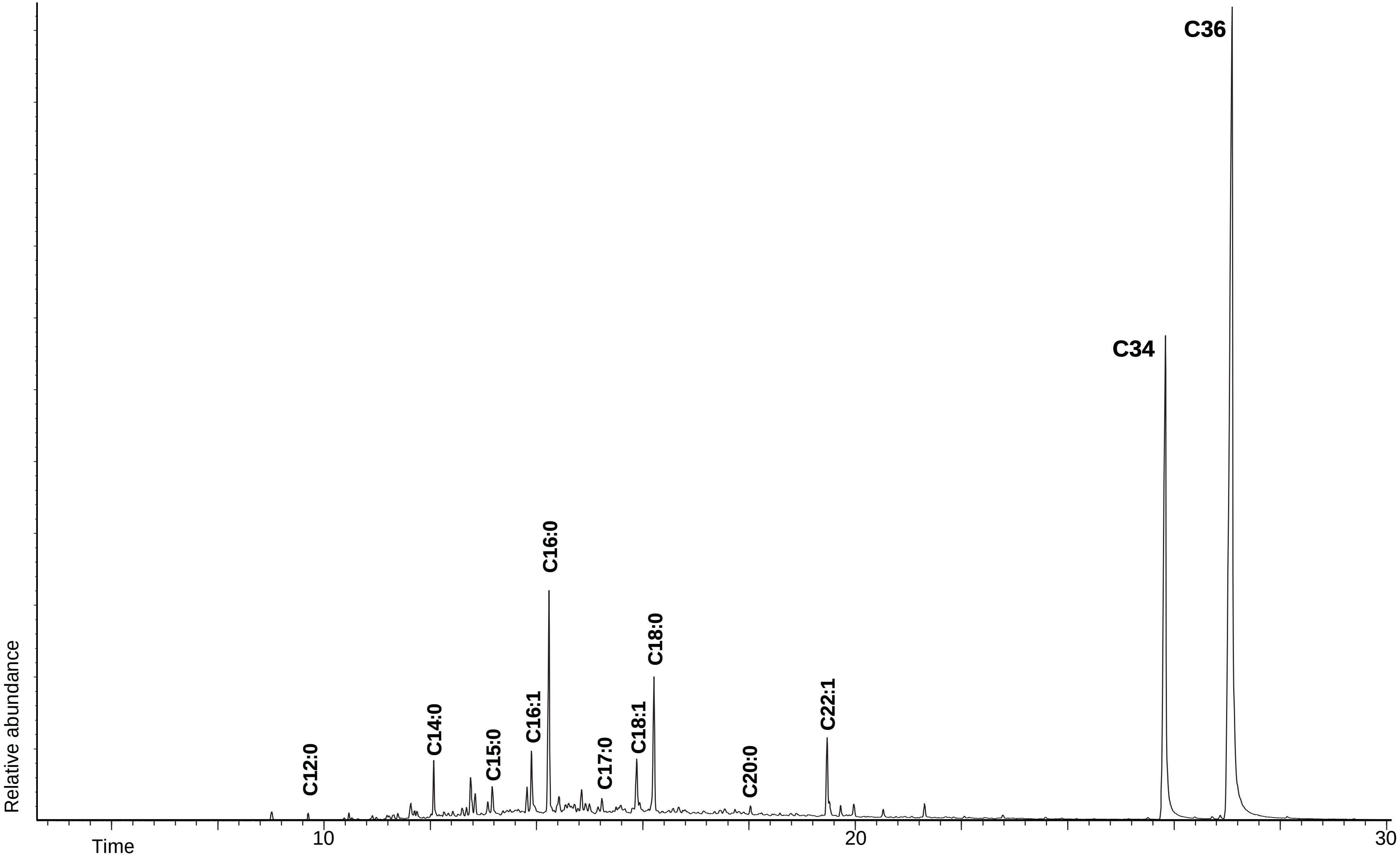
CSPF

C36
C34
C16:0
C18:0
C14:0
C15:0
C16:1
C12:0
C17:0
C18:1
C20:0
C22:1
Relative abundance
Time
10
20
30

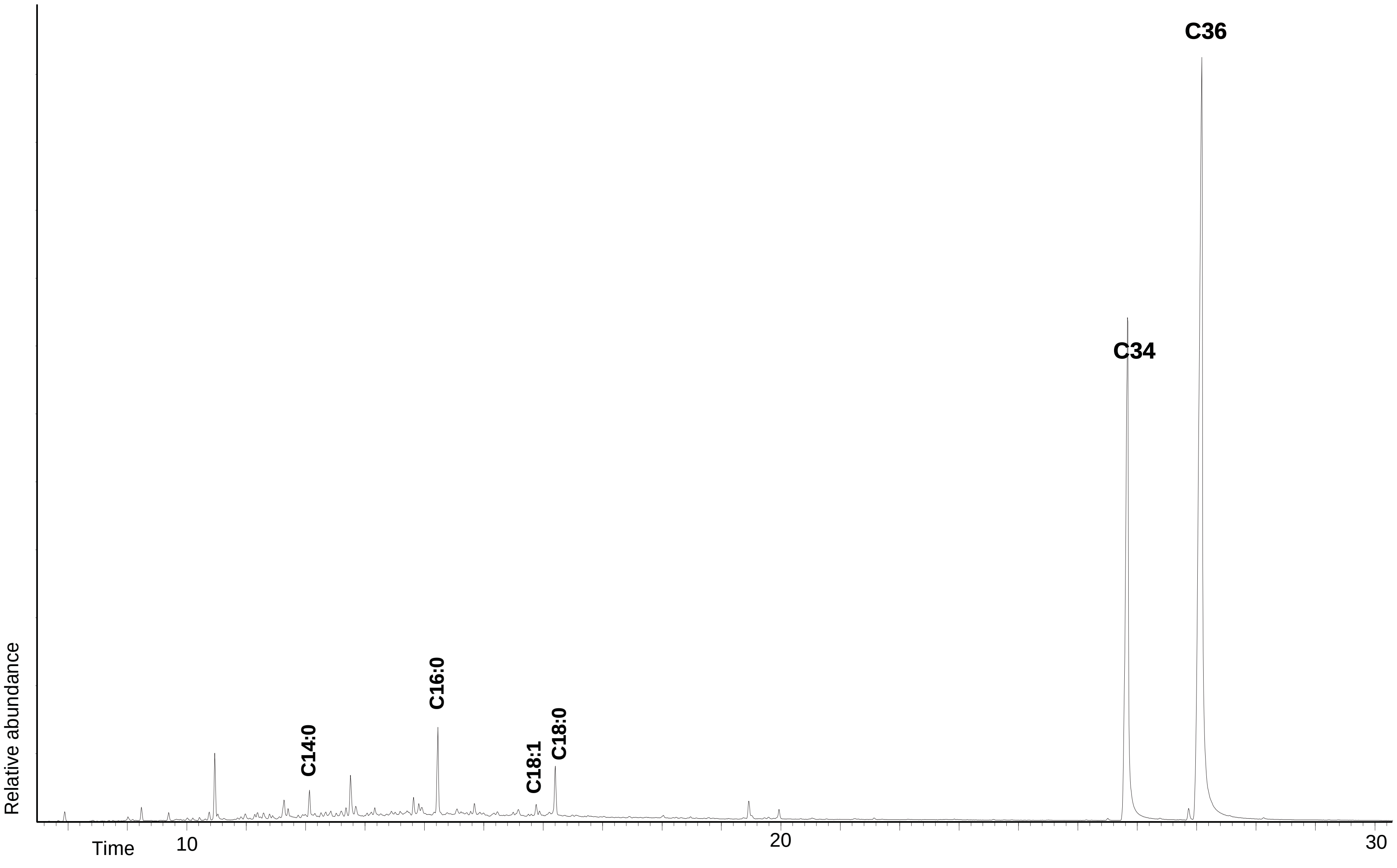
CSPG

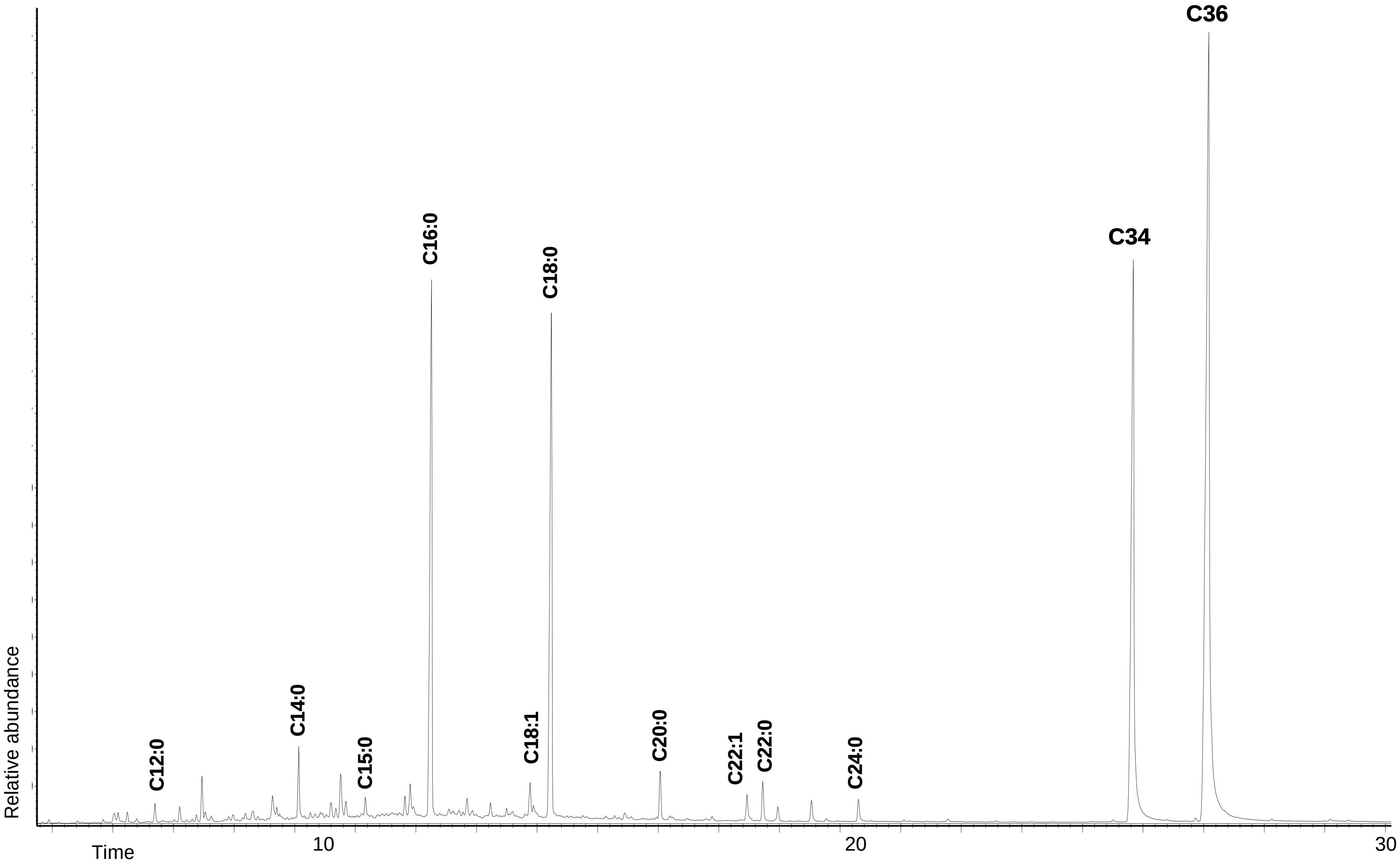

C36
C34
C16:0
C18:0
C12:0
C14:0
C15:0
C16:1
C17:0
C18:1
C20:0
C22:1
Relative abundance
Time
10
20
30

CSPH


Relative abundance
C12:0
C14:0
C15:0
C16:1
C16:0
C17:0
C18:1
C18:0
C20:0
C22:1
C34
C36
Time
10
20
30

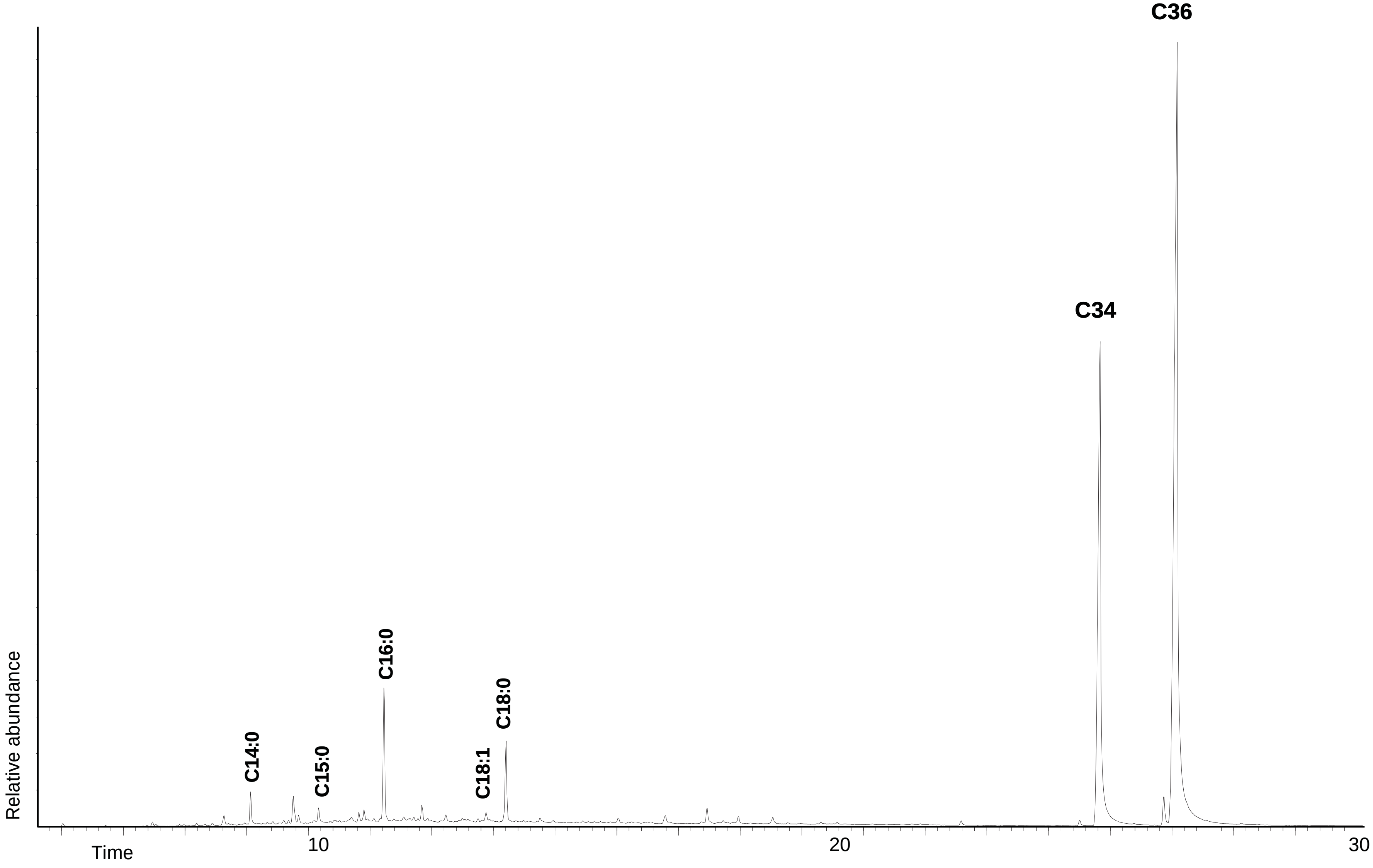
CSPI





CSPJ

C36
C34
C16:0
C18:0
C12:0
C14:0
C15:0
C18:1
C20:0
C22:1
C22:0
C24:0
Relative abundance
Time
10
20
30

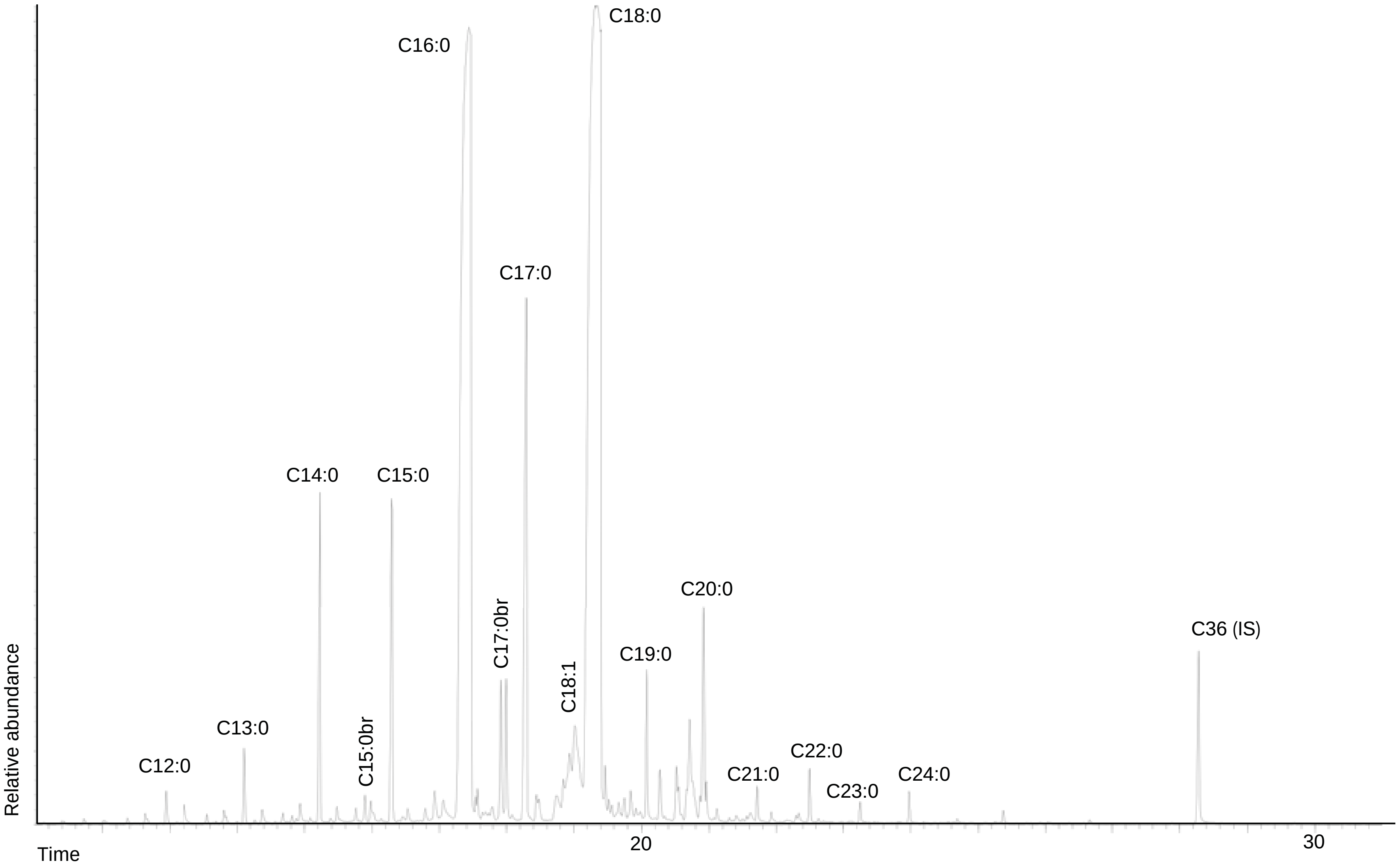


CSPK

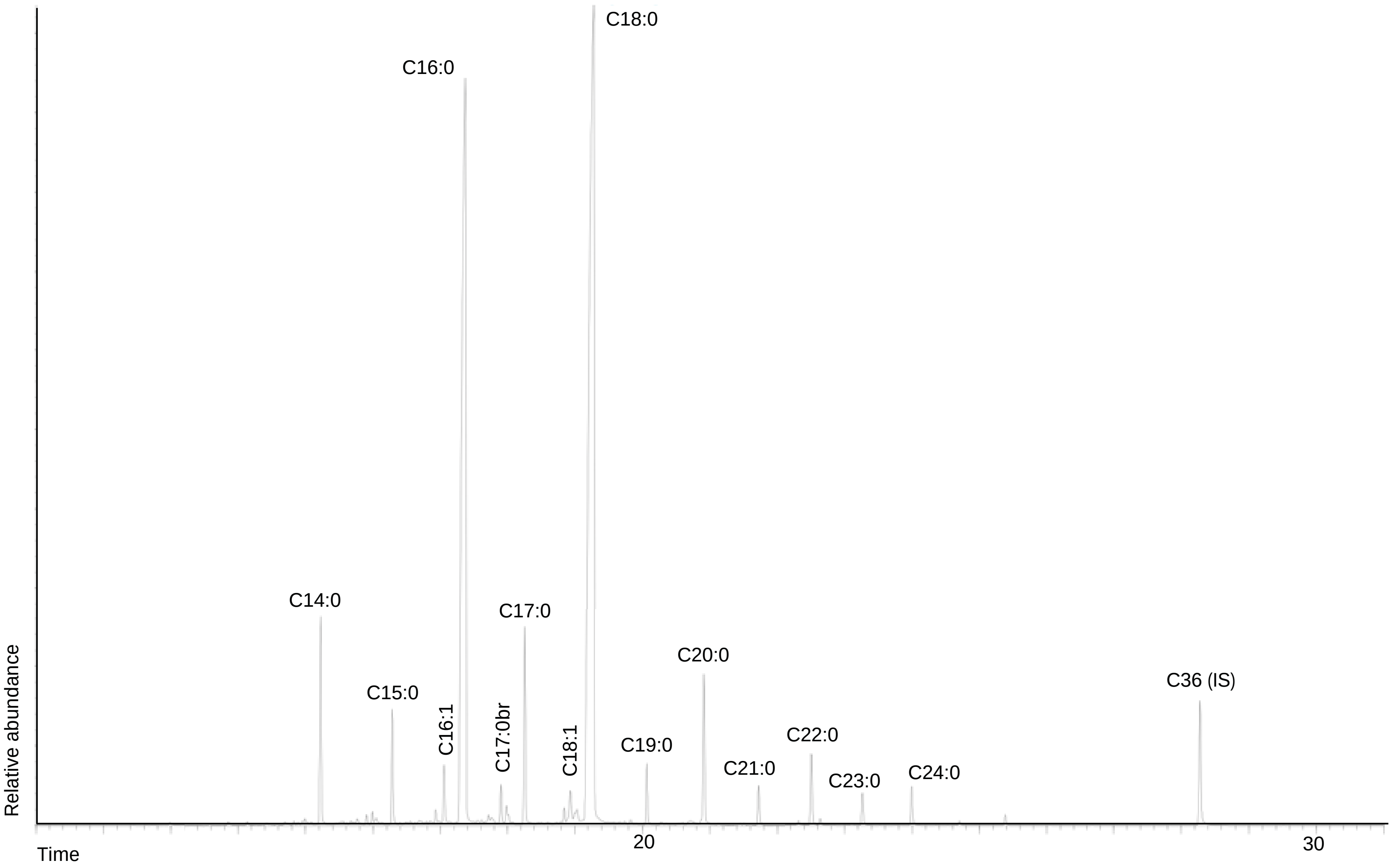
C36
C34
C16:0
C18:0
C14:0
C15:0
C18:1
Relative abundance
Time
10
20
30

Carrer Reina Amàlia

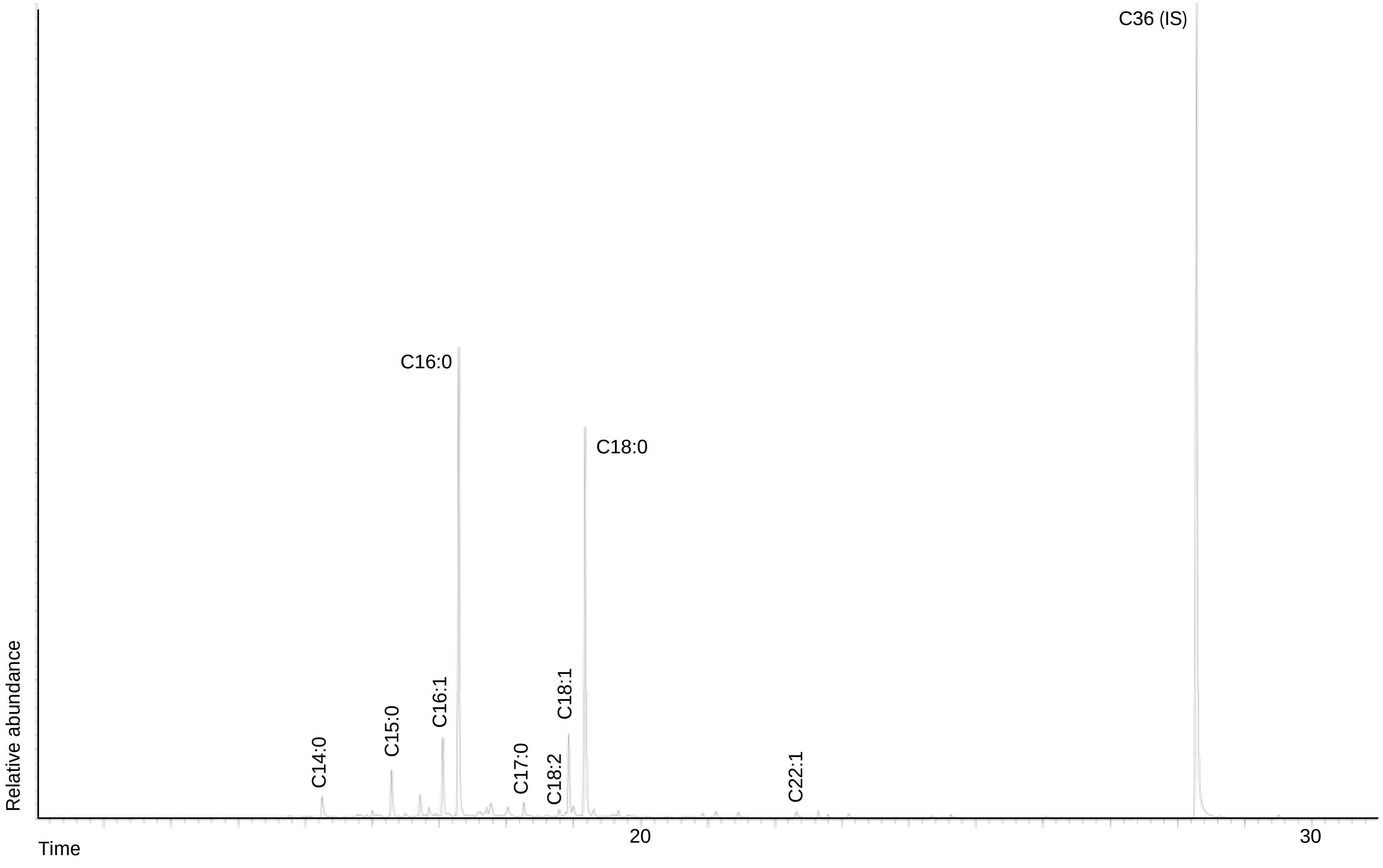
RA 6695


Relative abundance
Time
C12:0
C13:0
C14:0
C15:0br
C15:0
C16:0
C17:0br
C17:0
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C36 (IS)
20
30

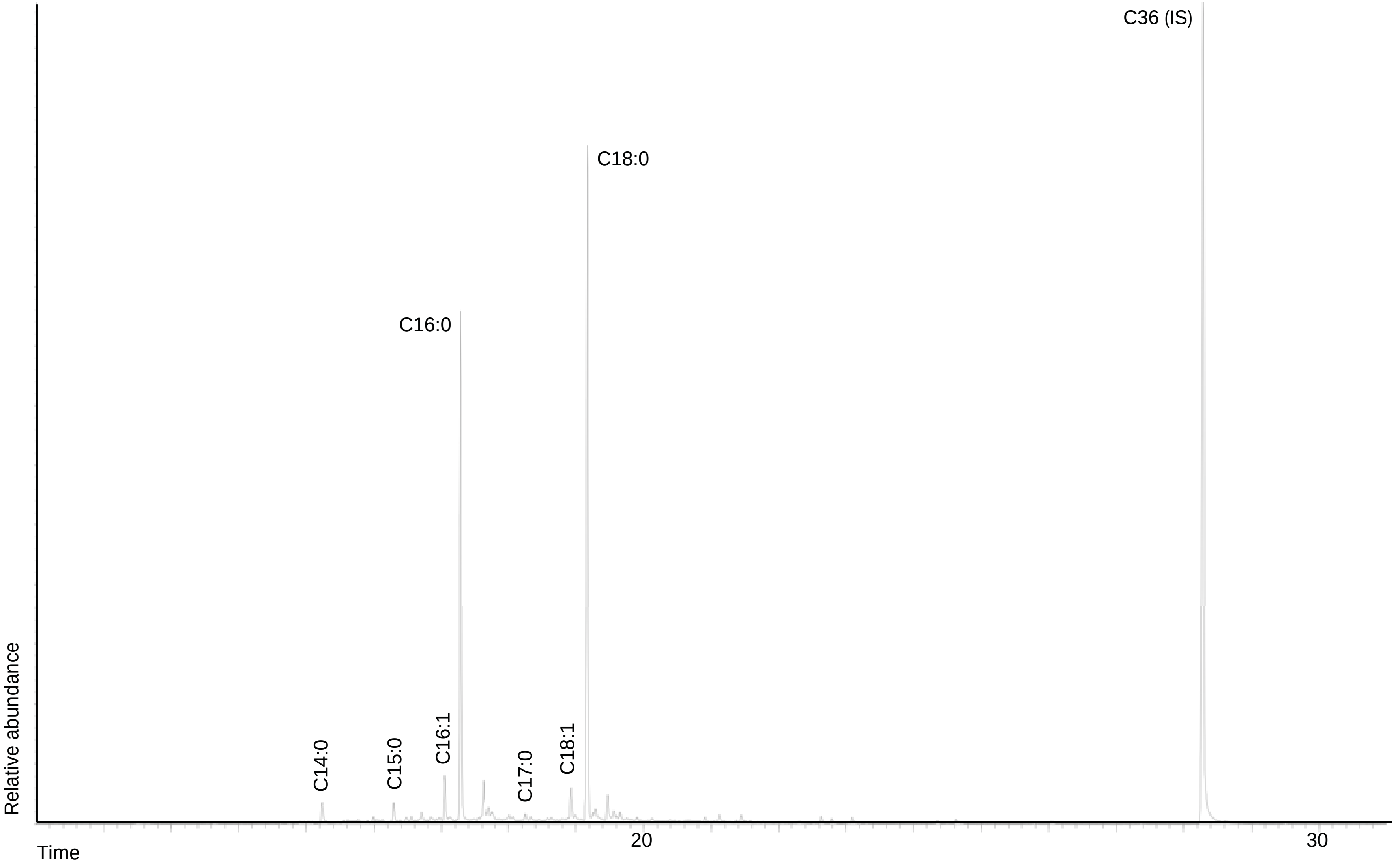
RA 6725


C18:0
C16:0
C14:0
C17:0
C15:0
C16:1
C17:0br
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C36 (IS)
Relative abundance
Time
20
30

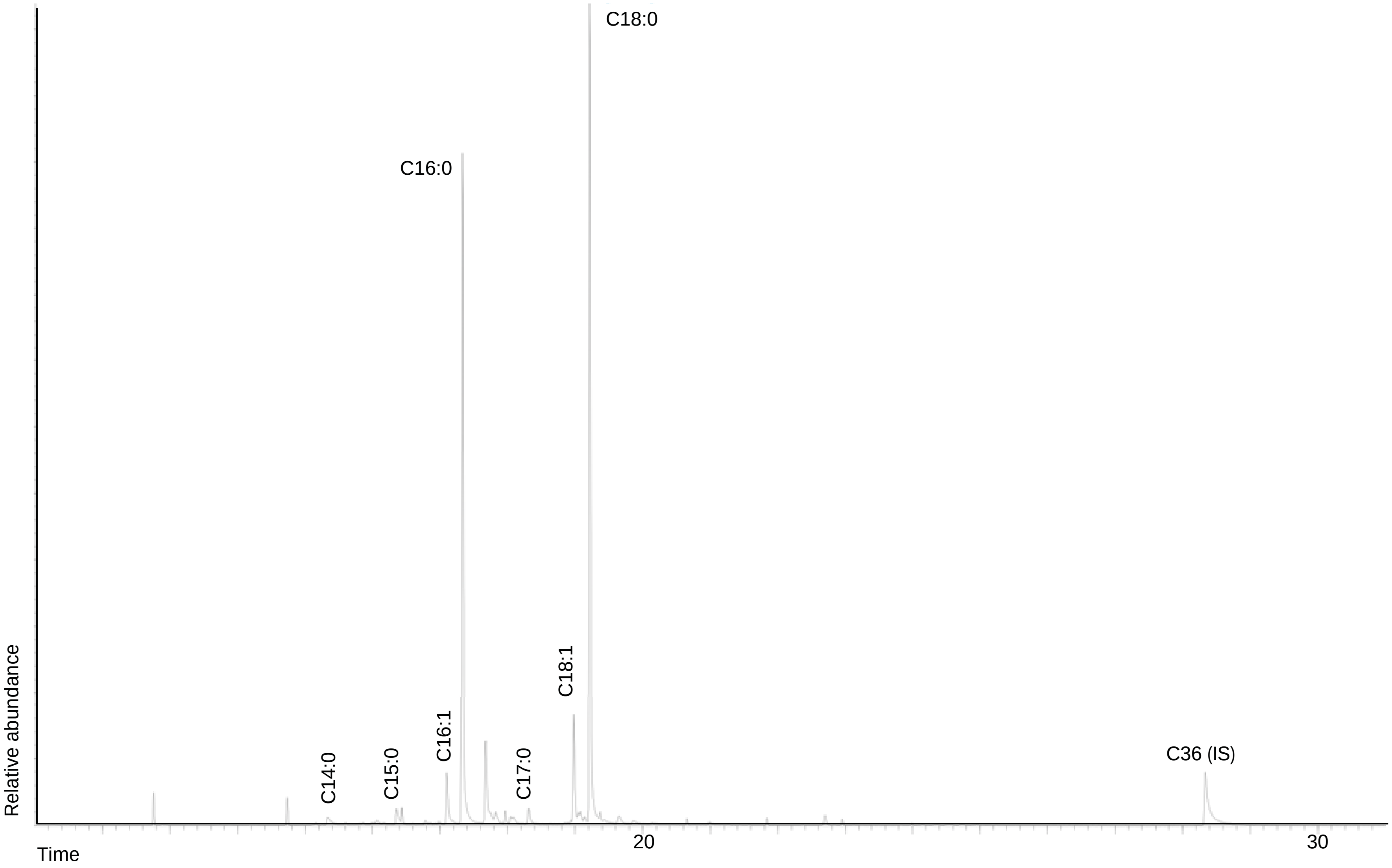
RA 7656


C36 (IS)
C16:0
C18:0
C14:0
C15:0
C16:1
C17:0
C18:2
C18:1
C22:1
Relative abundance
Time
20
30

RA 7718

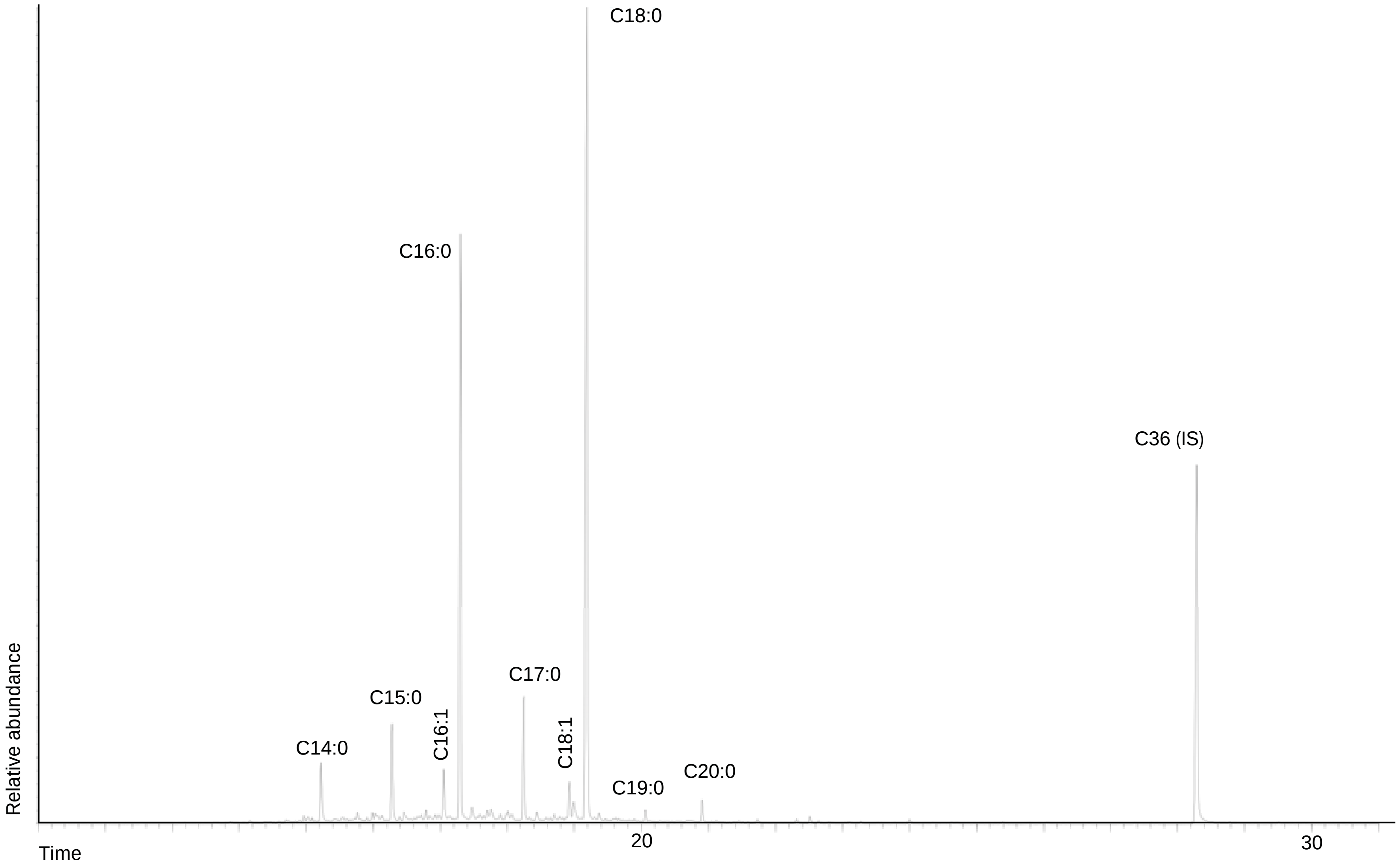

C36 (IS)
C18:0
C16:0
C14:0
C15:0
C16:1
C17:0
C18:1
Relative abundance
Time
20
30

RA 7778

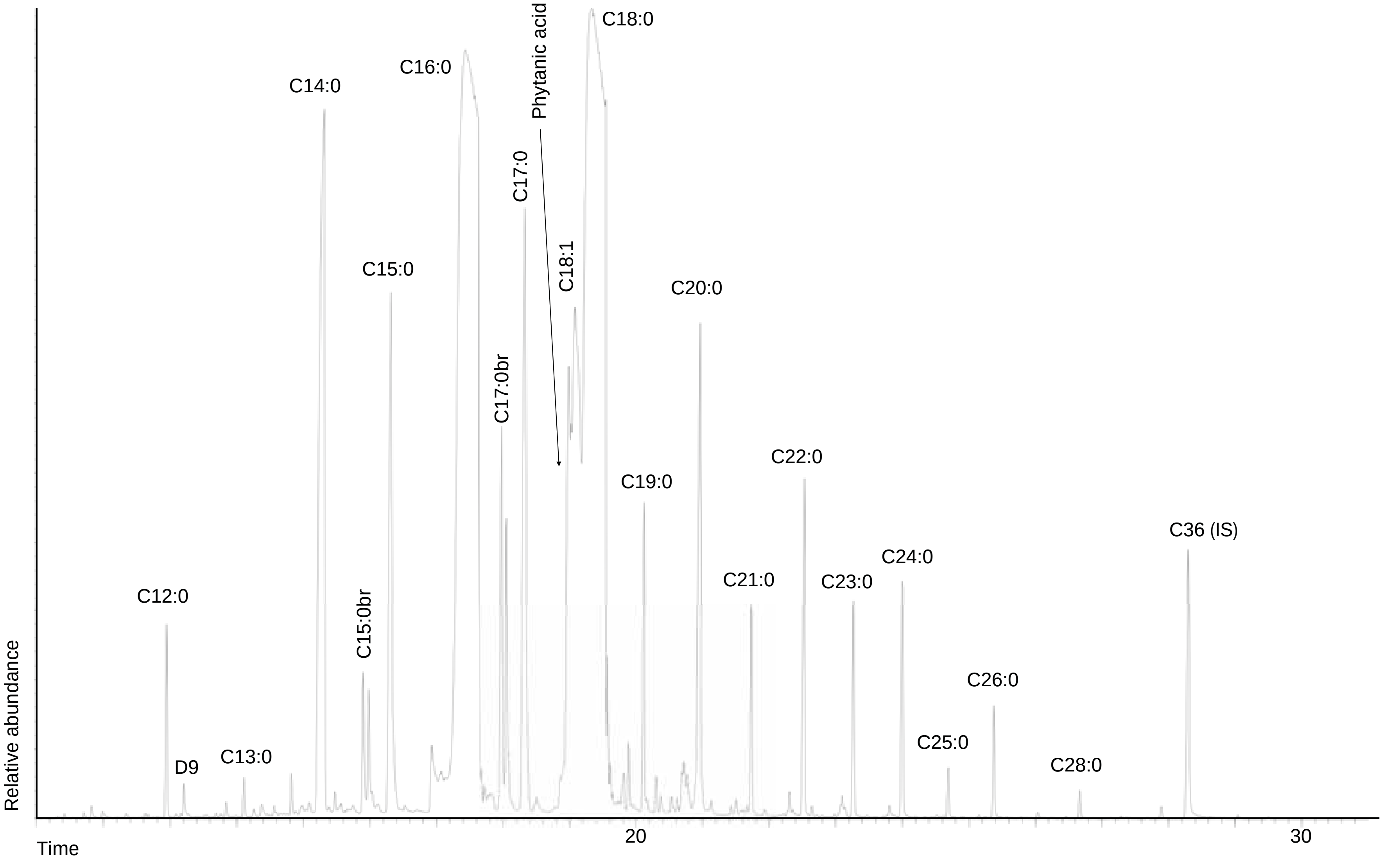


C18:0
C16:0
C18:1
C16:1
C14:0
C15:0
C17:0
C36 (IS)
Relative abundance
Time
20
30

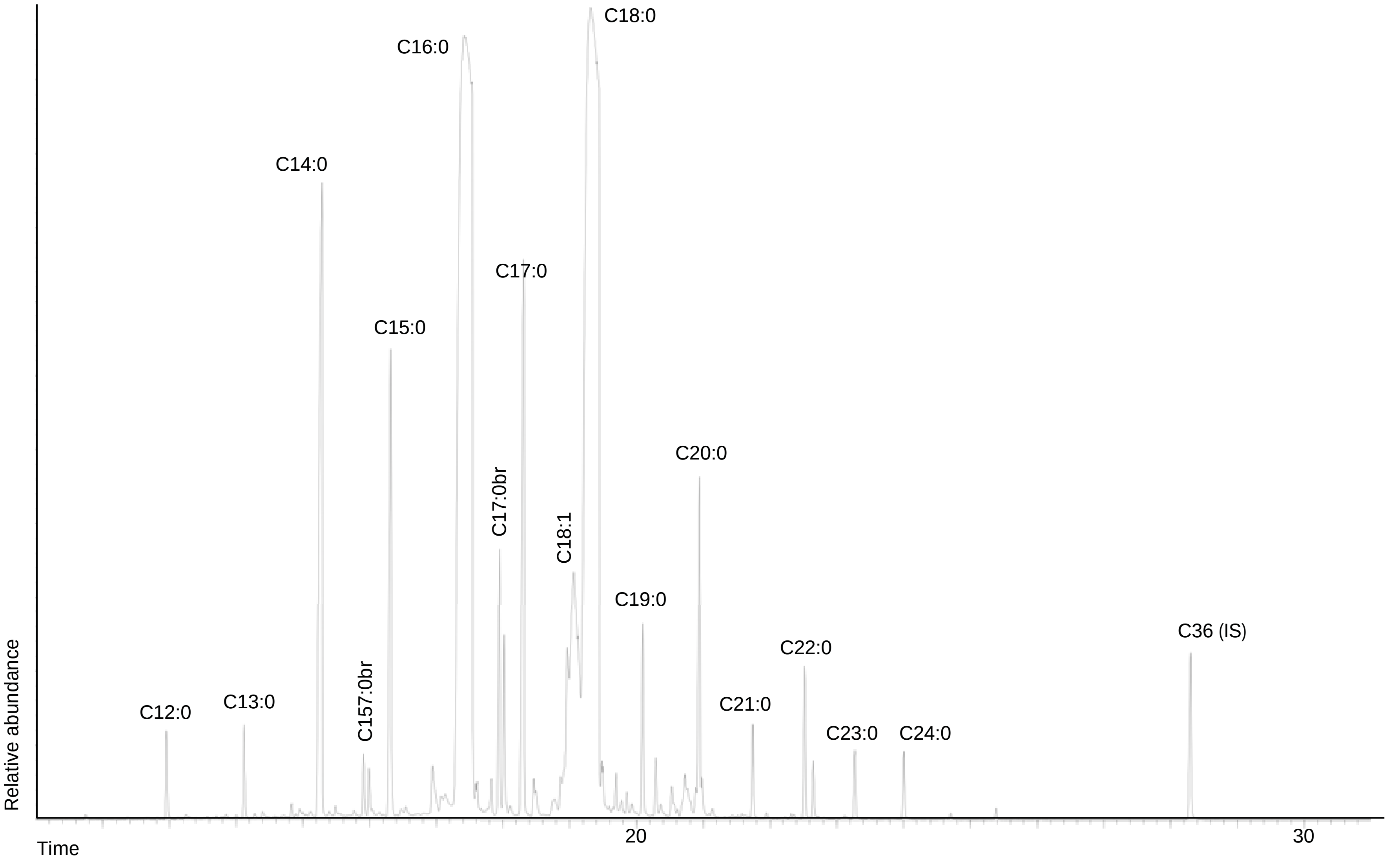
RA 8009


C18:0
C16:0
C36 (IS)
C17:0
C15:0
C14:0
C16:1
C18:1
C19:0
C20:0
Relative abundance
Time
20
30

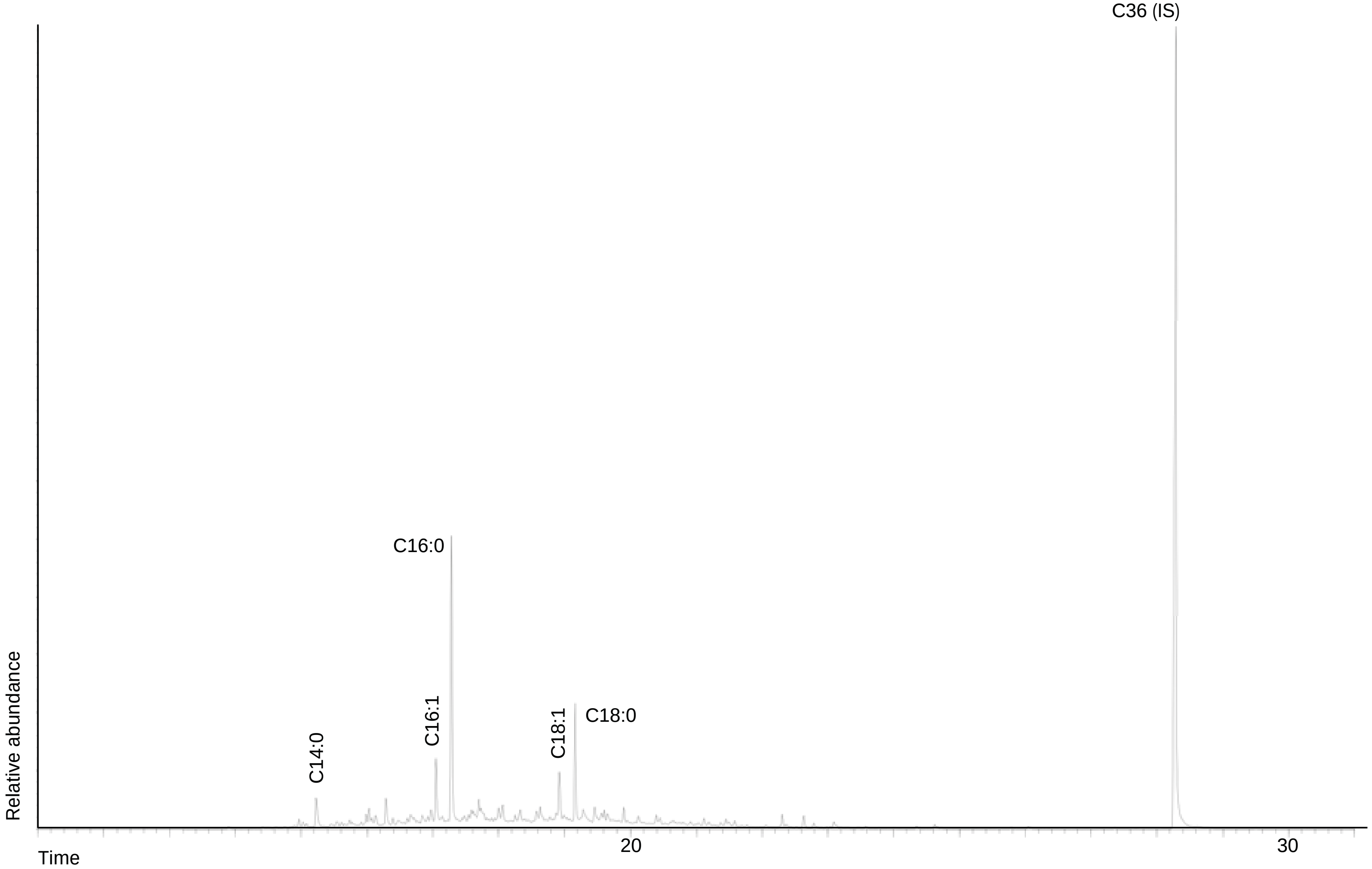
RA 8070


C18:0
Phytanic acid
C16:0
C14:0
C17:0
C18:1
C15:0
C20:0
C17:0br
C22:0
C19:0
C36 (IS)
C24:0
C21:0
C23:0
C12:0
C15:0br
C26:0
C25:0
C28:0
C13:0
D9
Relative abundance
Time
20
30

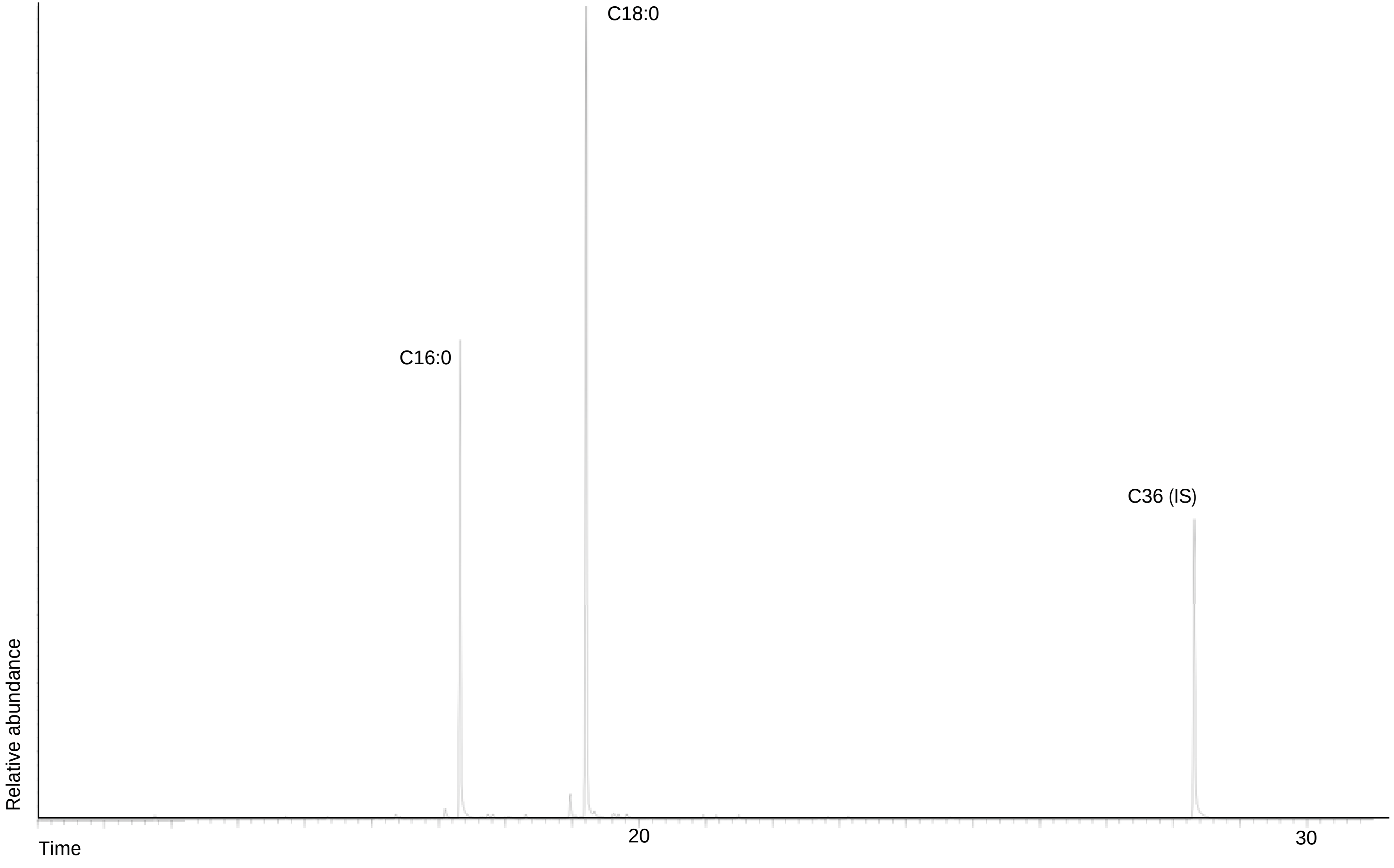
RA 8234


C18:0
C16:0
C14:0
C17:0
C15:0
C20:0
C17:0br
C18:1
C19:0
C36 (IS)
C22:0
C12:0
C13:0
C157:0br
C21:0
C23:0
C24:0
Relative abundance
Time
20
30

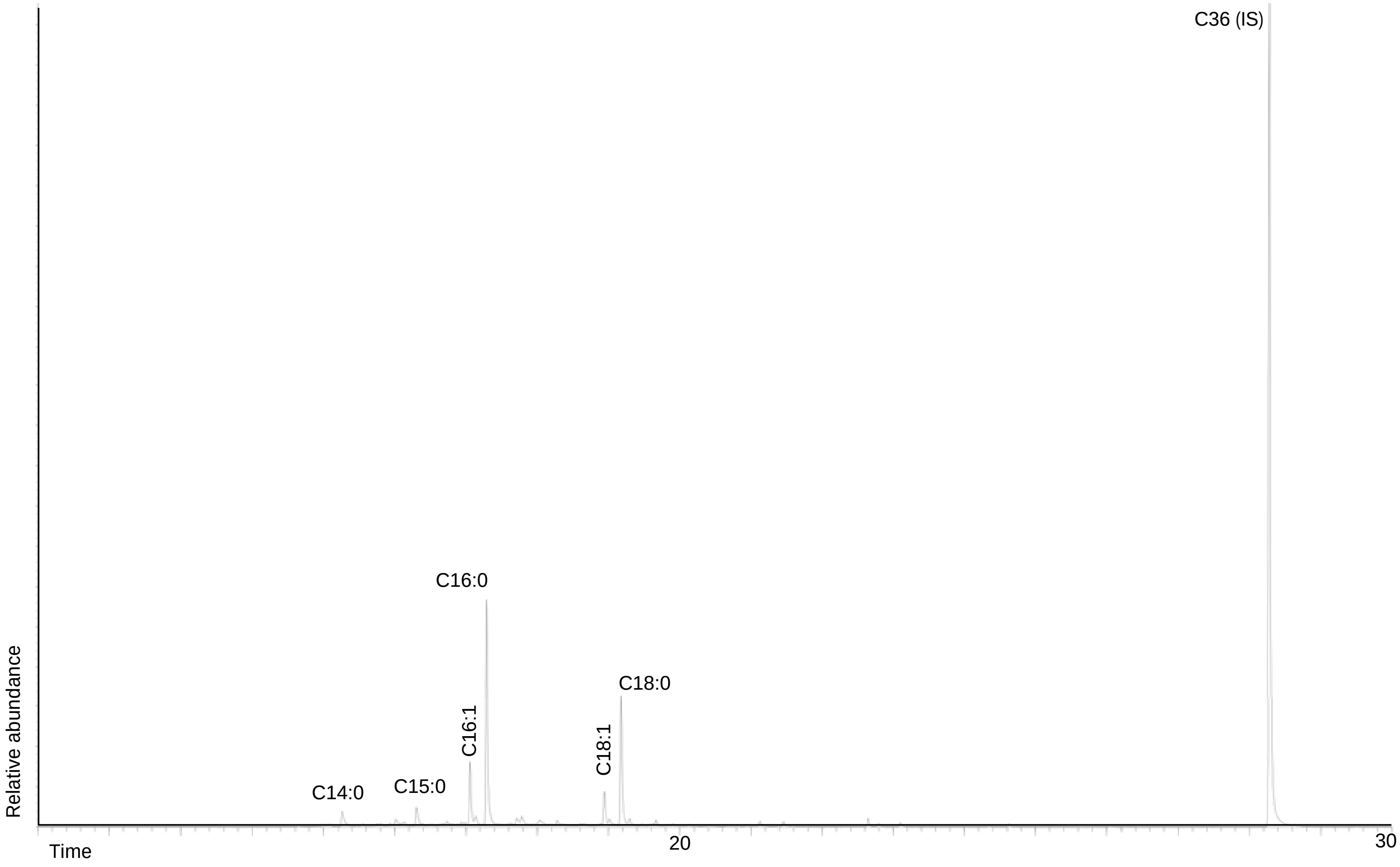
RA 8263


C36 (IS)
C16:0
C16:1
C14:0
C18:1
C18:0
Relative abundance
Time
20
30

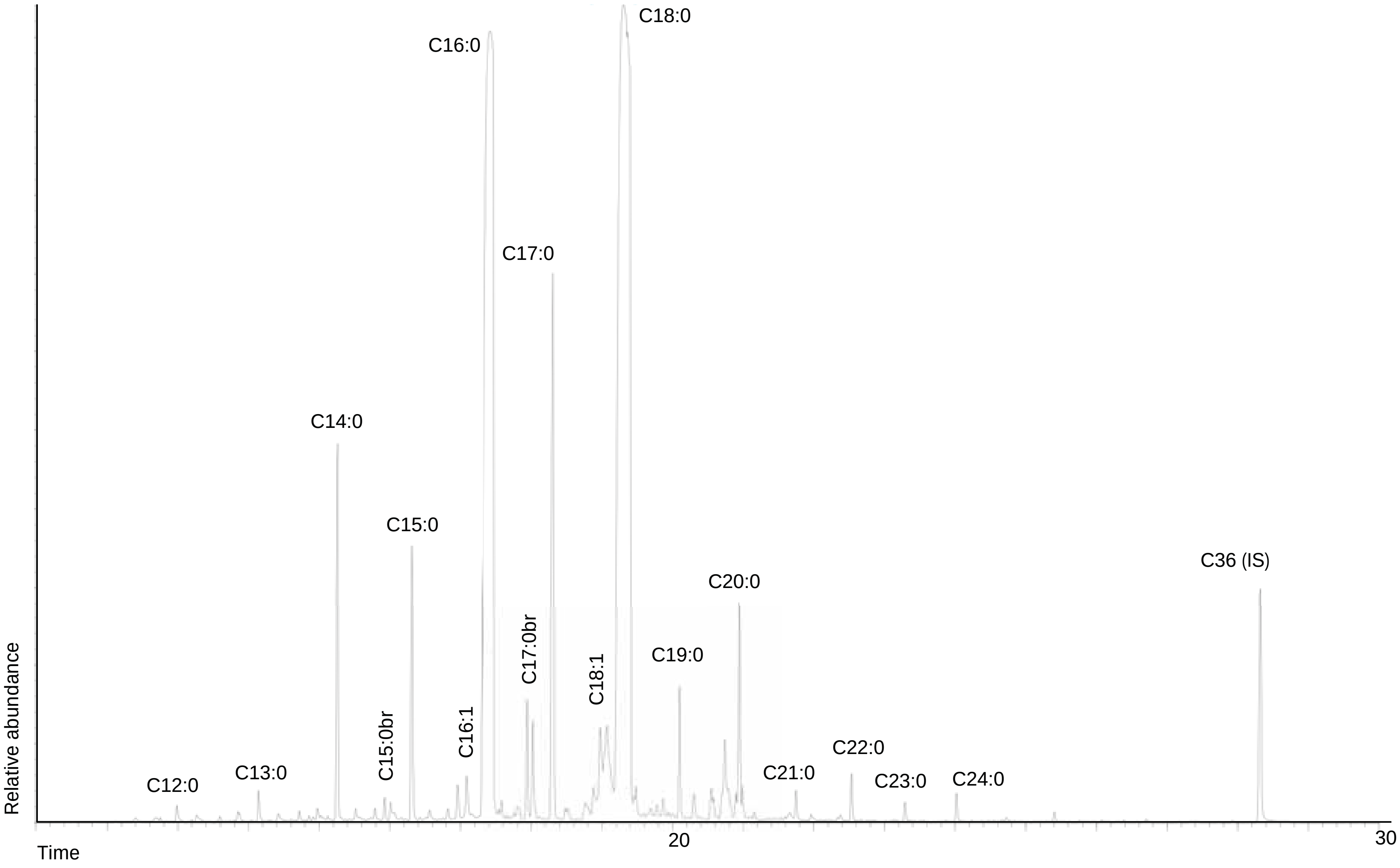
RA 8392



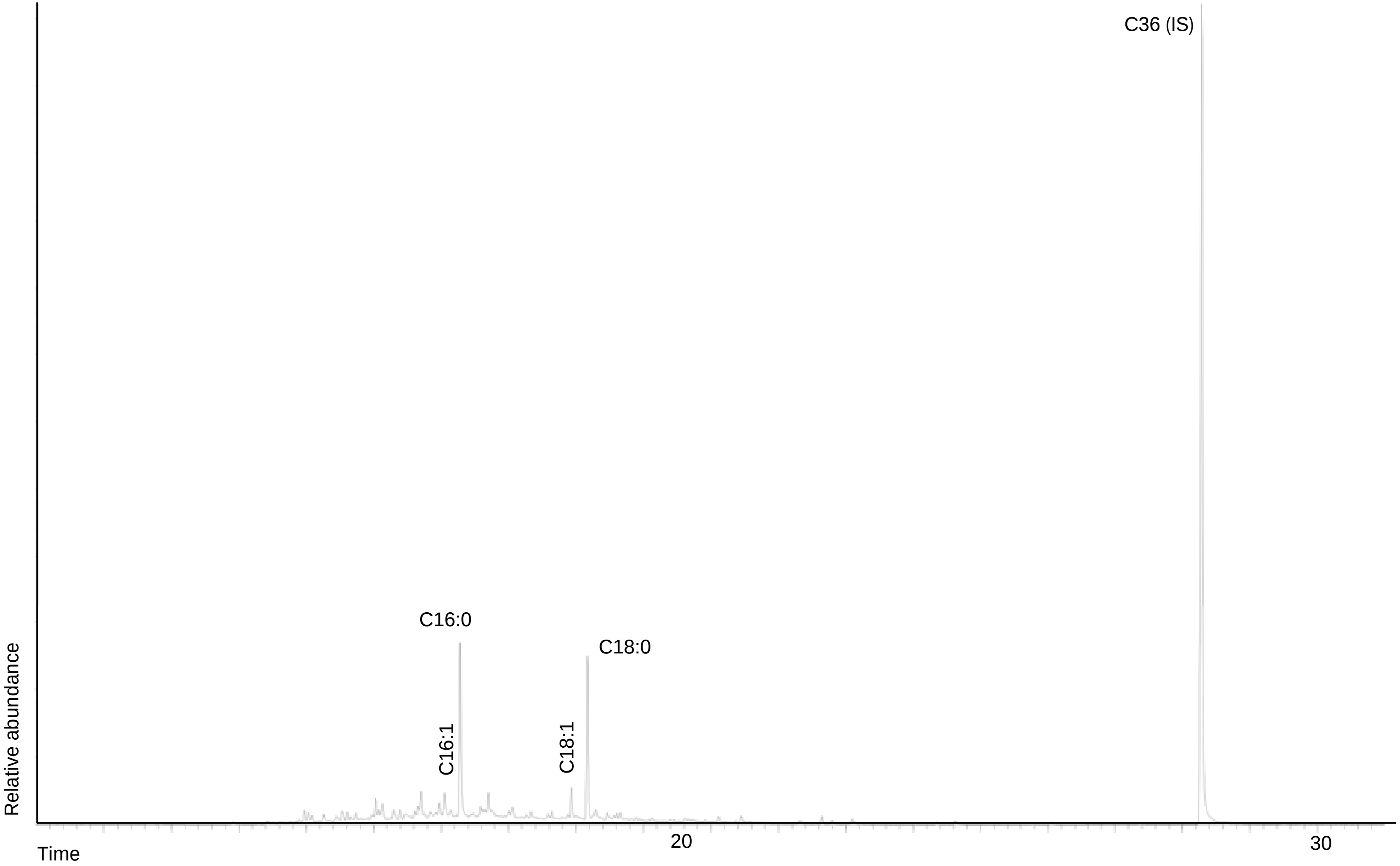
RA 4892


C36 (IS)
C16:0
C18:0
C16:1
C18:1
C14:0
C15:0
Relative abundance
Time
20
30

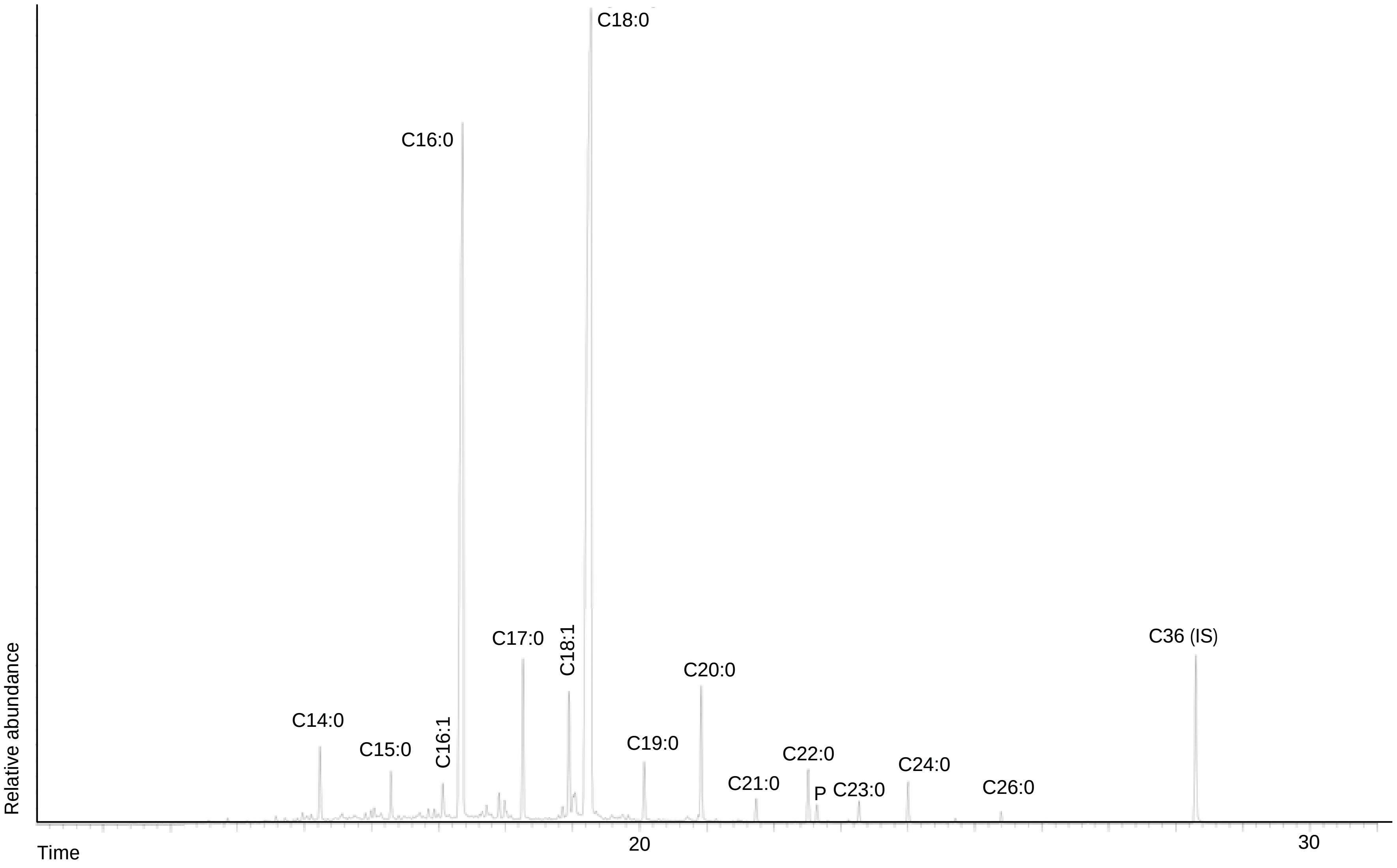
RA 5531


C18:0
C16:0
C17:0
C14:0
C15:0
C36 (IS)
C20:0
C17:0br
C19:0
C18:1
C15:0br
C16:1
C22:0
C12:0
C13:0
C21:0
C23:0
C24:0
Relative abundance
Time
20
30

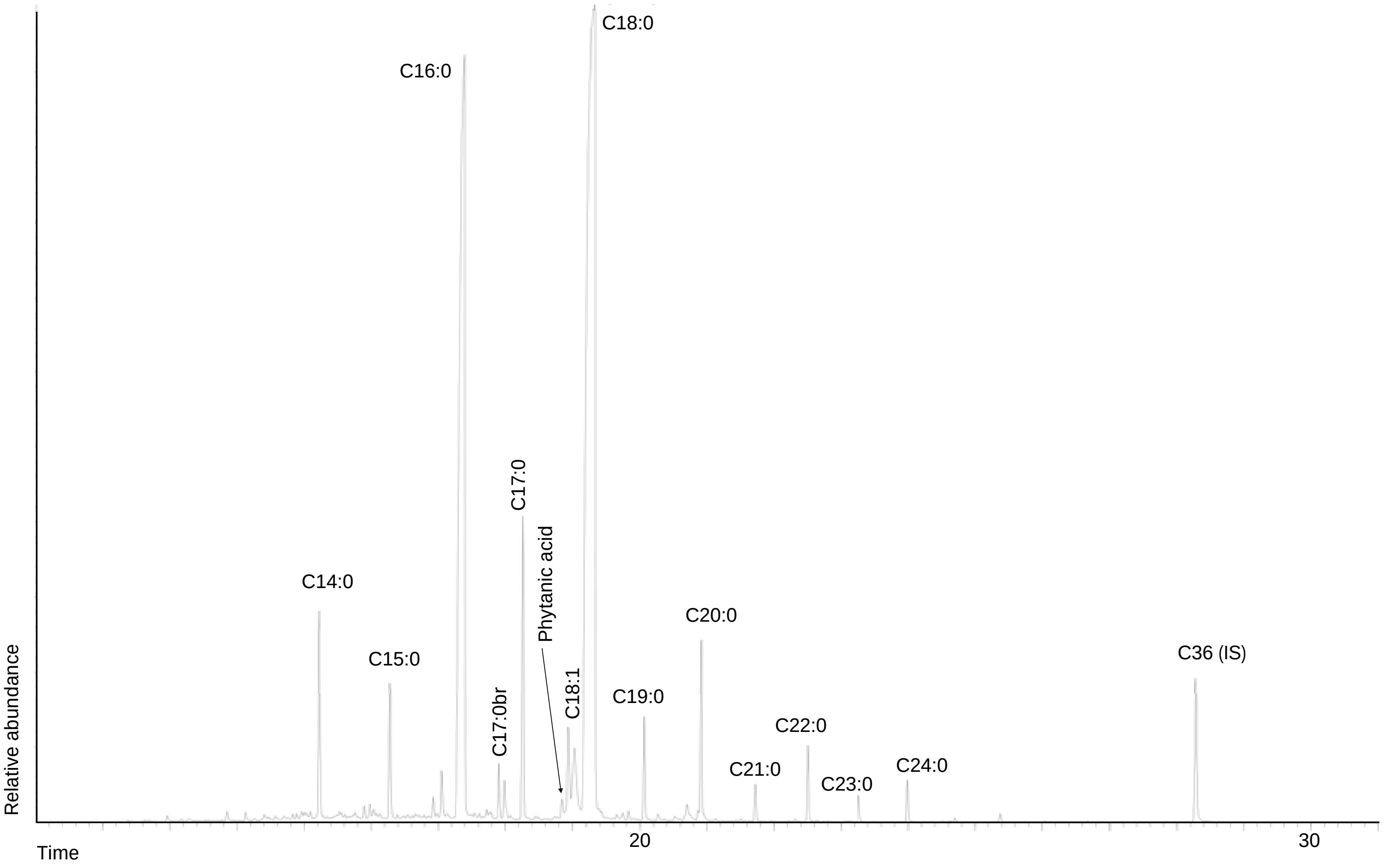
RA 5561



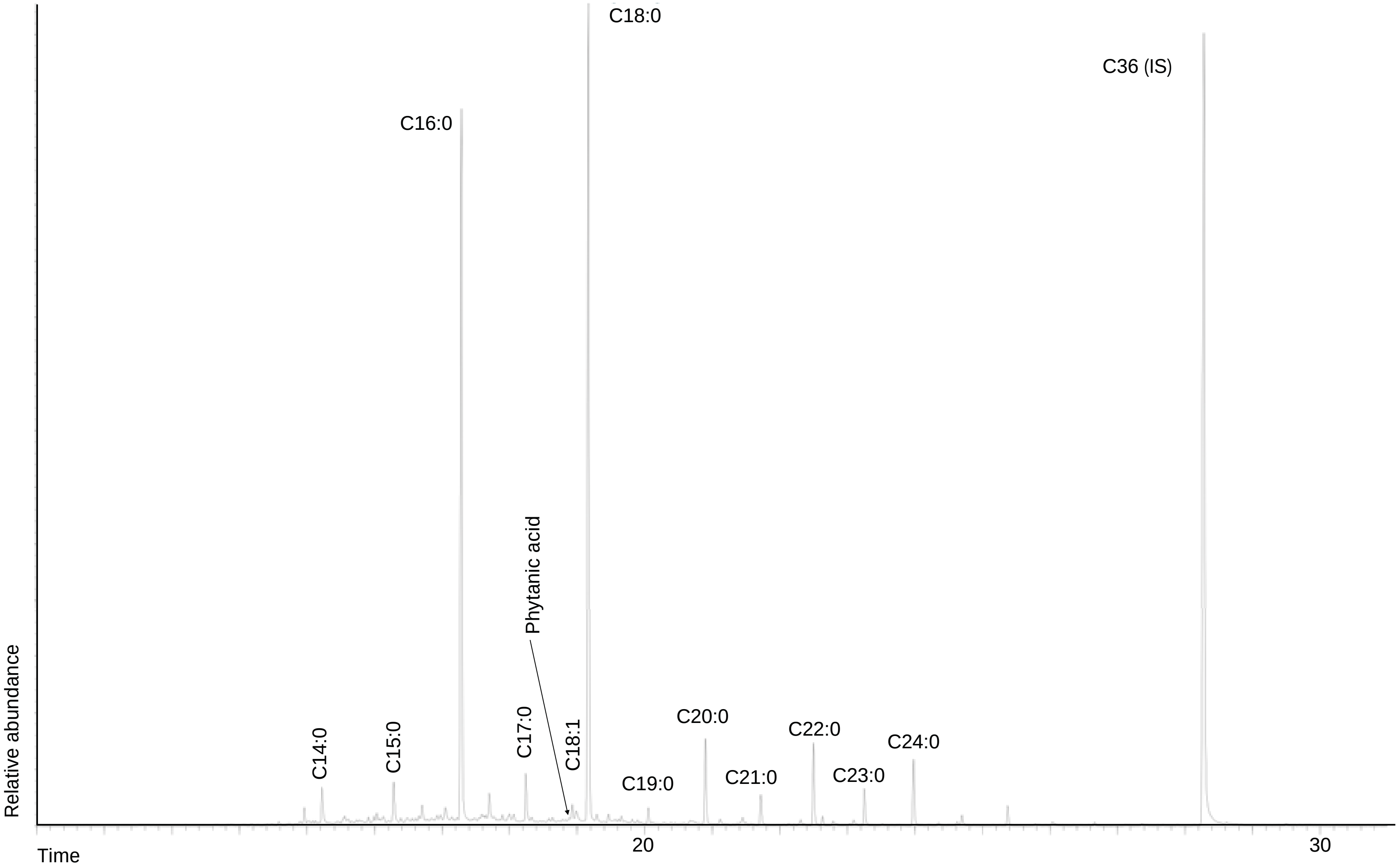
RA 5823


C18:0
C16:0
C17:0
C18:1
C36 (IS)
C20:0
C14:0
C19:0
C15:0
C16:1
C22:0
C24:0
C21:0
C26:0
P
C23:0
Relative abundance
Time
20
30

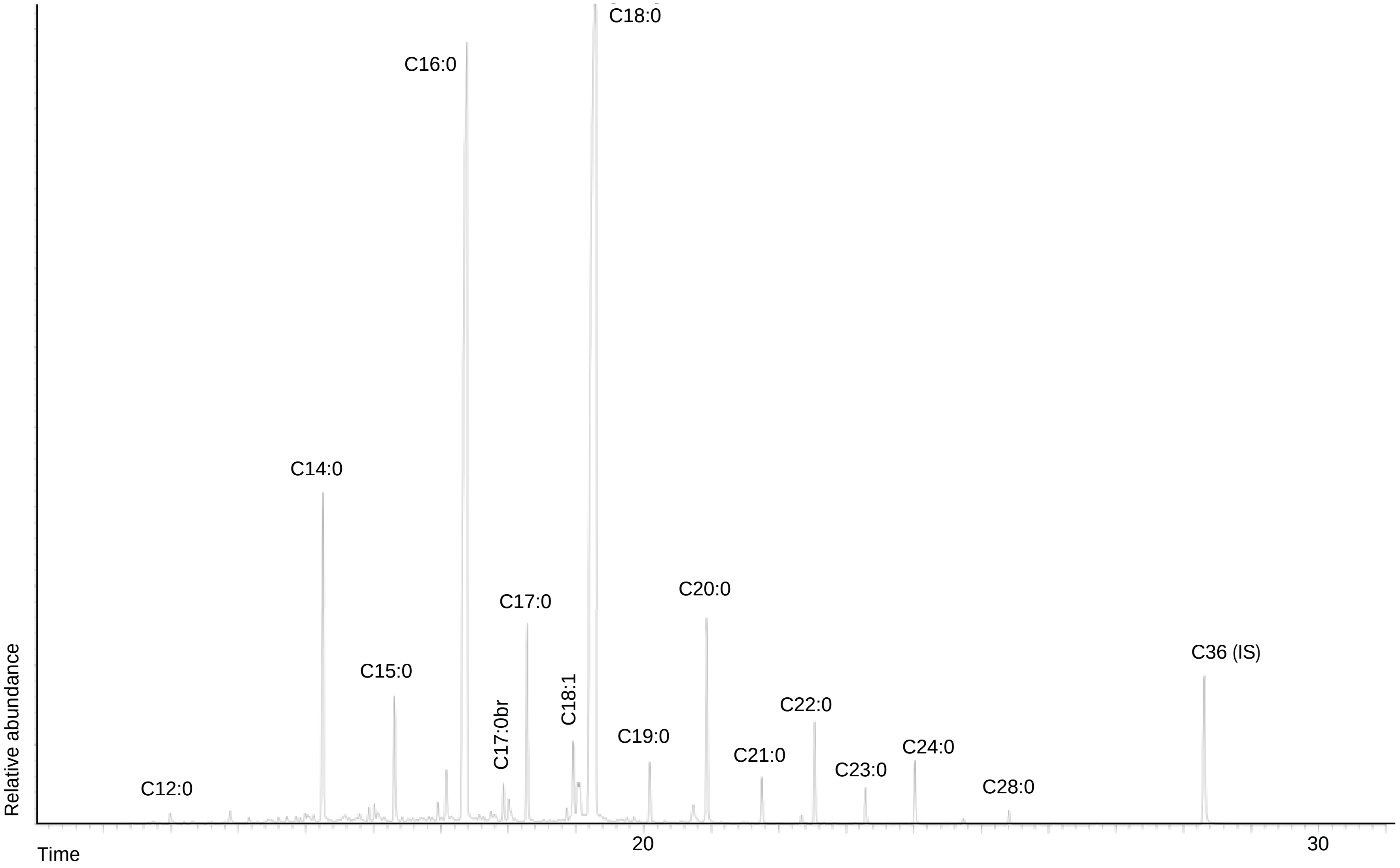
RA 6100


C18:0
C16:0
C17:0
Phytanic acid
C14:0
C20:0
C36 (IS)
C15:0
C18:1
C17:0br
C19:0
C22:0
C21:0
C24:0
C23:0
Relative abundance
Time
20
30

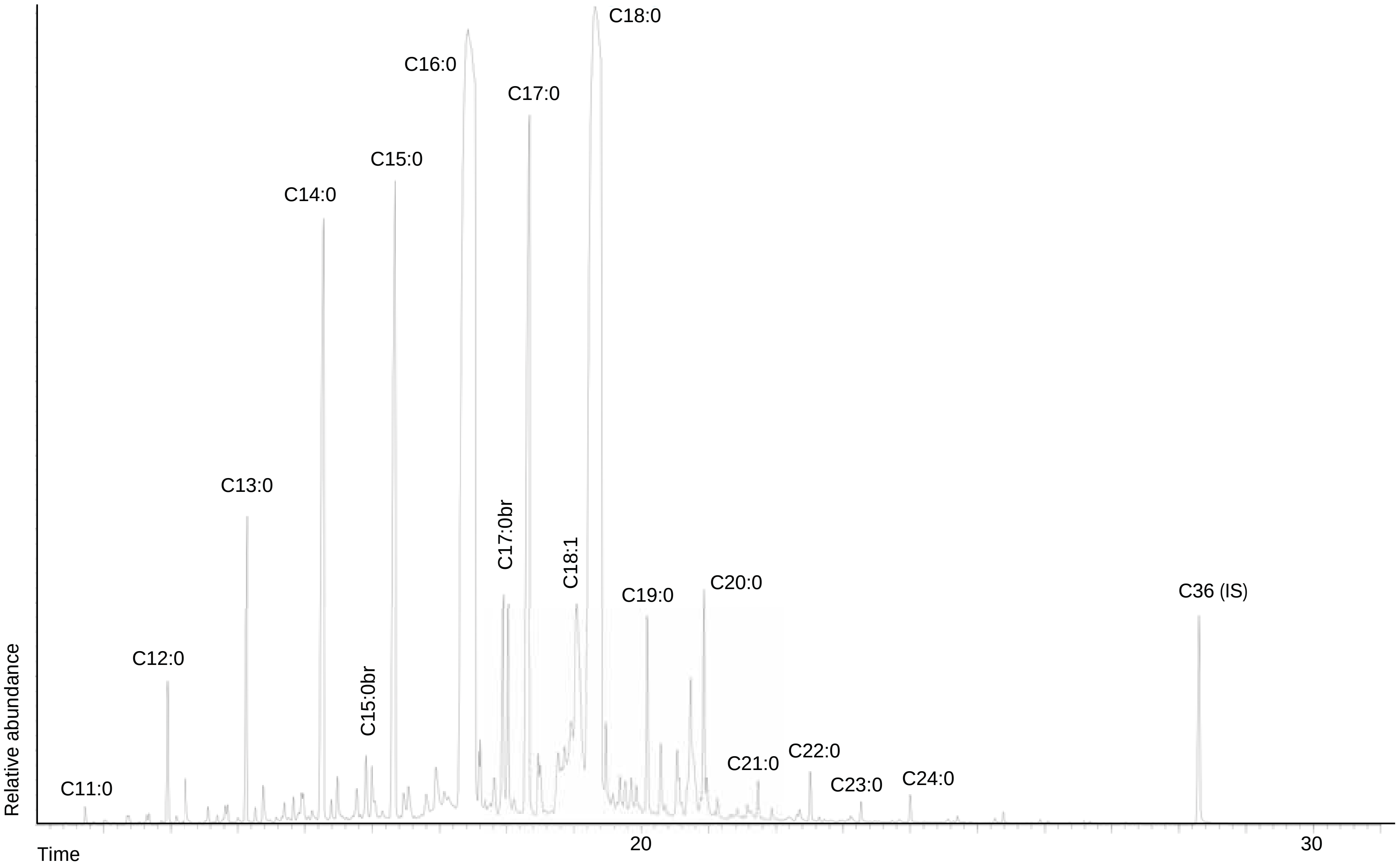
RA 6162


C18:0
C36 (IS)
C16:0
Phytanic acid
C14:0
C15:0
C17:0
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
Relative abundance
Time
20
30

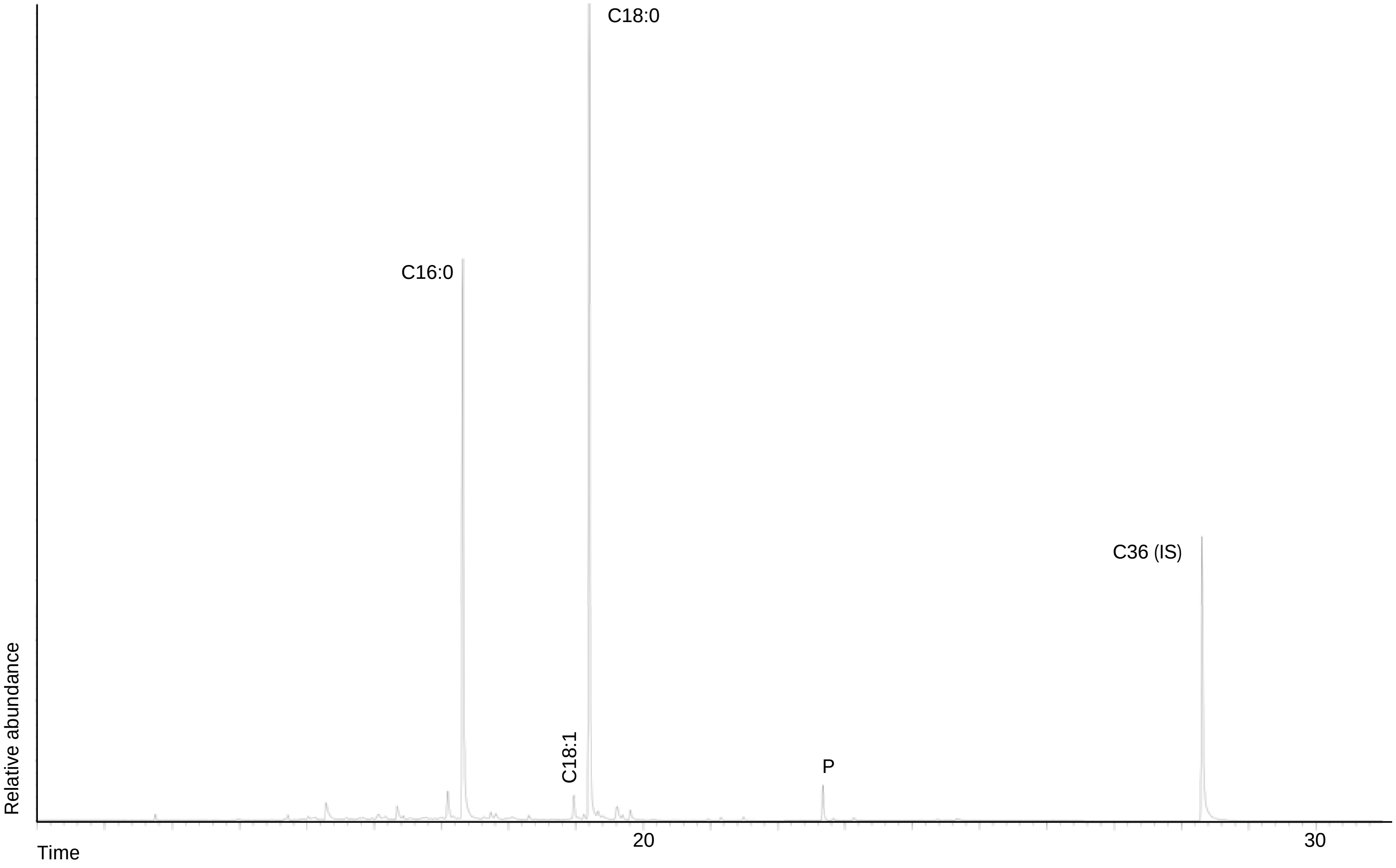
RA 6231


C18:0
C16:0
C14:0
C17:0
C20:0
C15:0
C36 (IS)
C17:0br
C18:1
C22:0
C19:0
C24:0
C21:0
C23:0
C12:0
C28:0
Relative abundance
20
30
Time

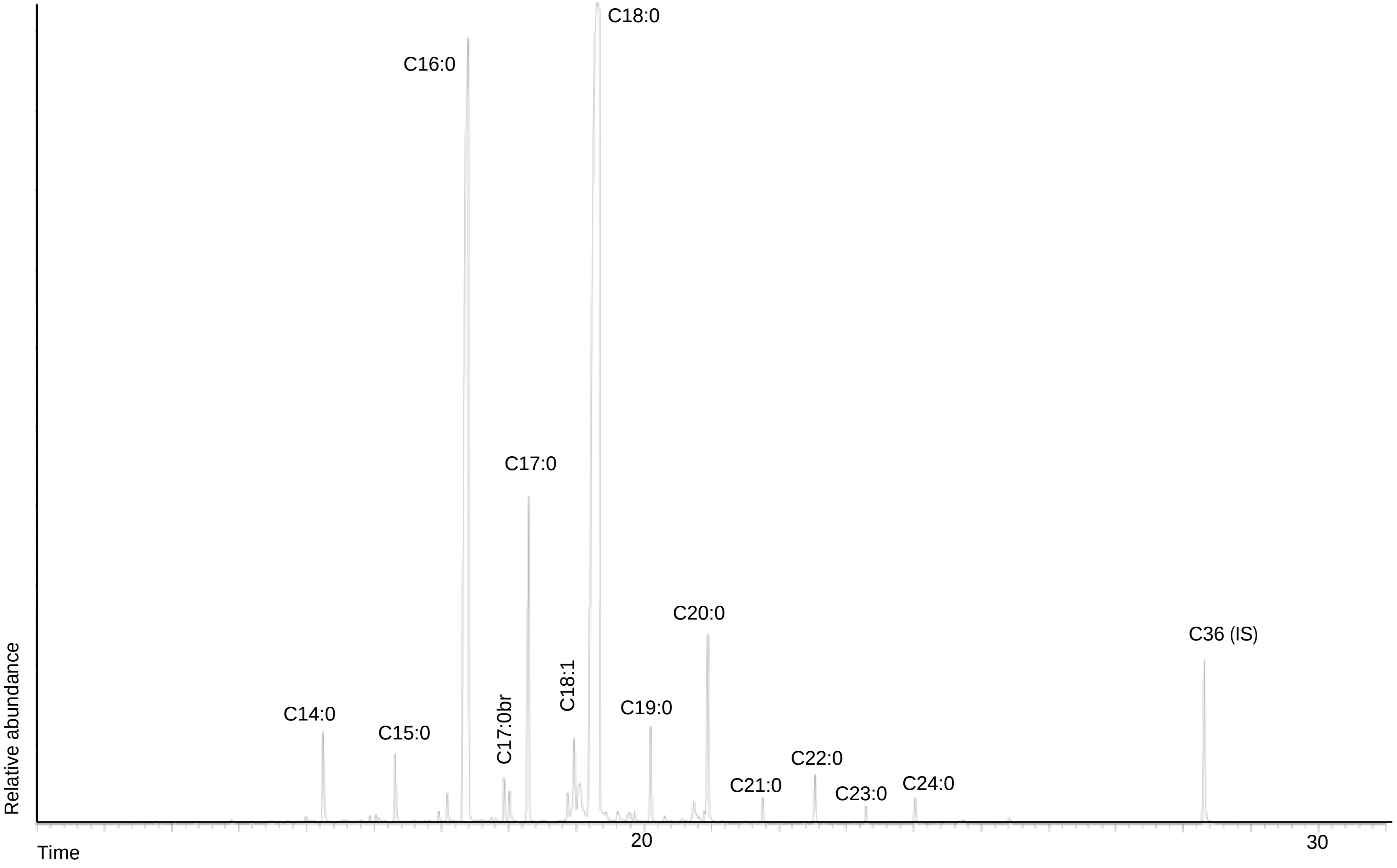
RA 6405


Relative abundance
C11:0
C12:0
C13:0
C14:0
C15:0br
C15:0
C16:0
C17:0br
C17:0
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C36 (IS)
Time
20
30

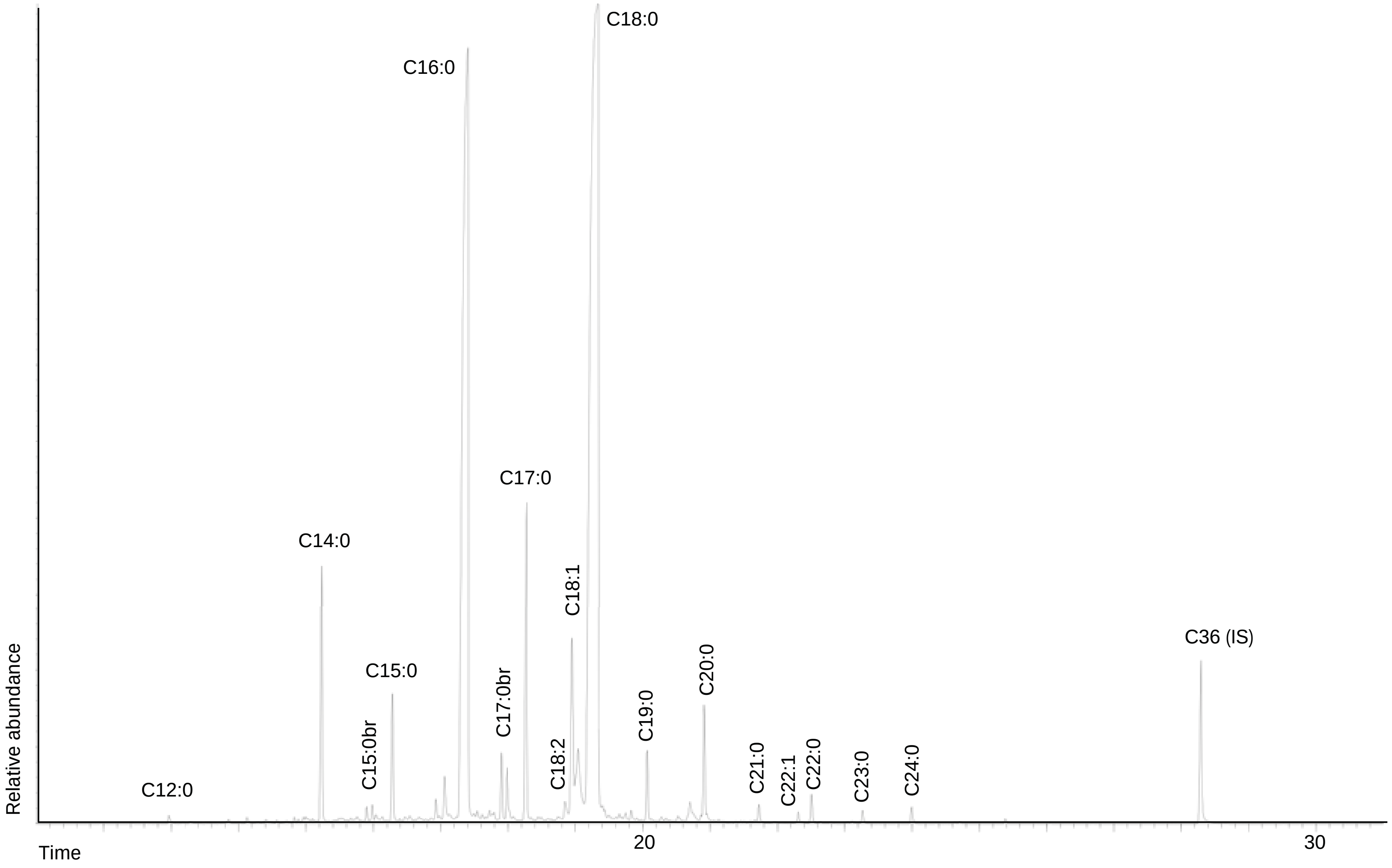
RA 6522



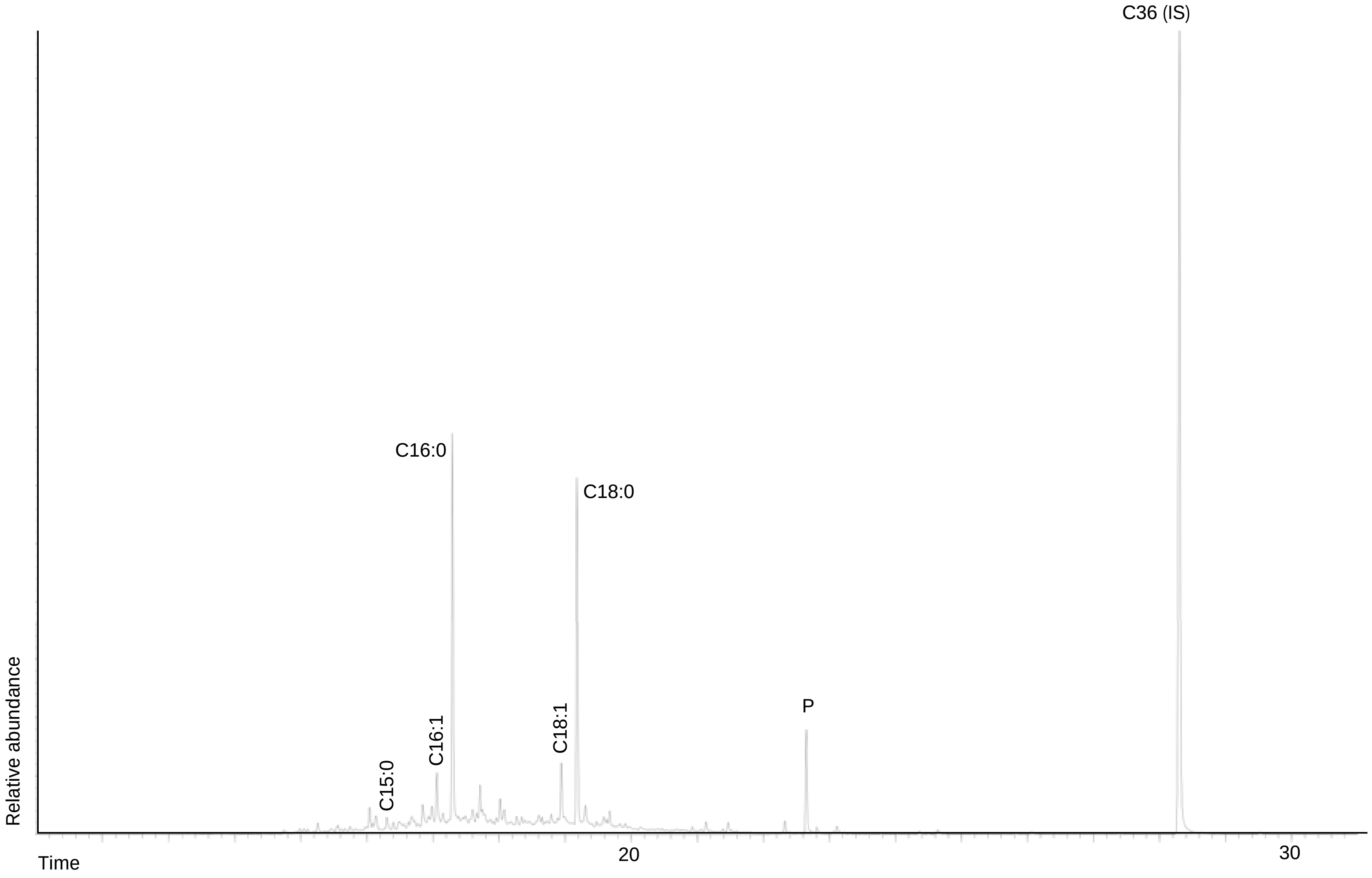
RA 6681


C18:0
C16:0
C17:0
C20:0
C36 (IS)
C18:1
C17:0br
C14:0
C19:0
C15:0
C22:0
C21:0
C24:0
C23:0
Relative abundance
Time
20
30

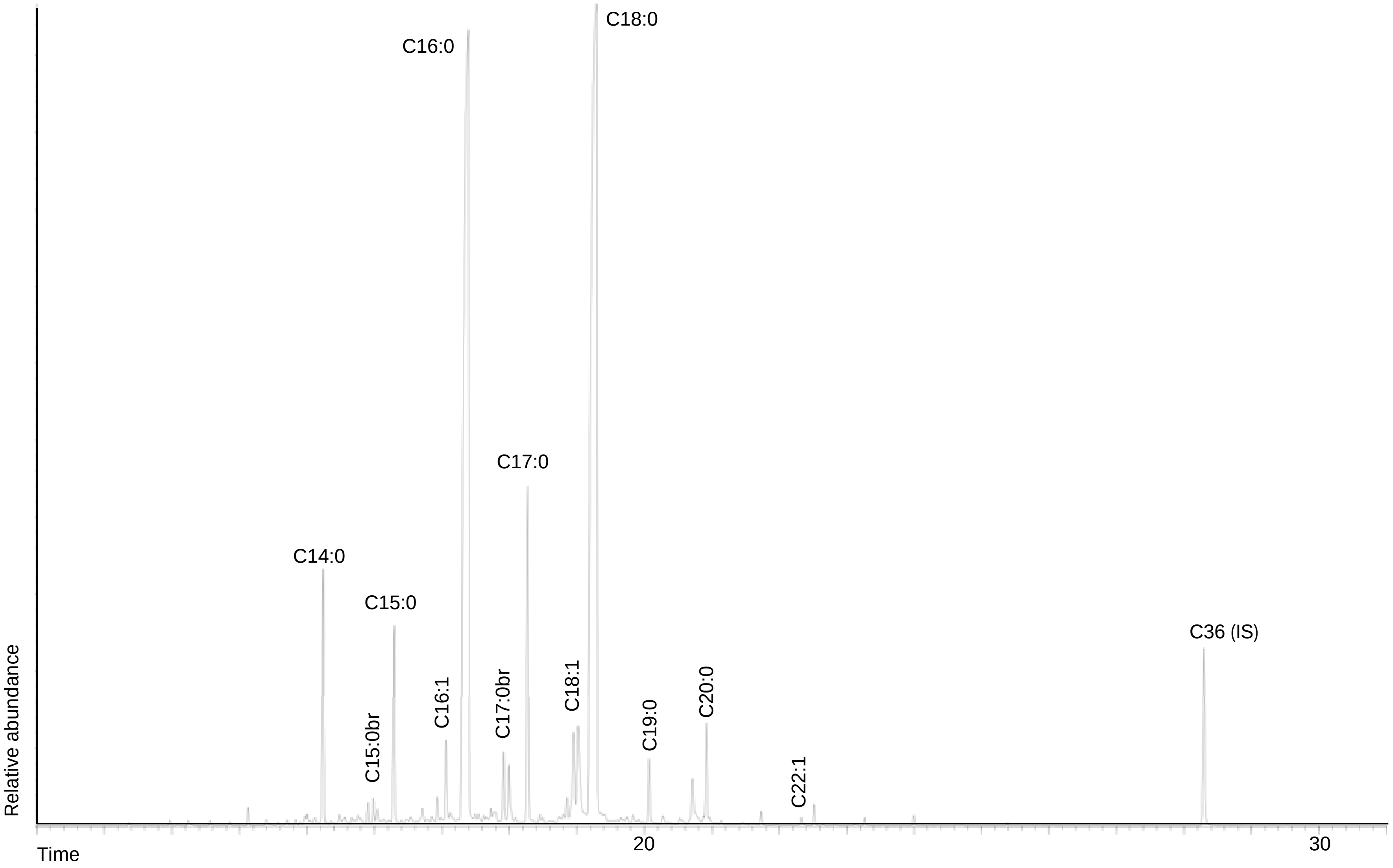
RA 8413


C18:0
C16:0
C17:0
C14:0
C18:1
C36 (IS)
C15:0
C20:0
C17:0br
C19:0
C15:0br
C18:2
C21:0
C22:1
C22:0
C23:0
C24:0
C12:0
Relative abundance
Time
20
30

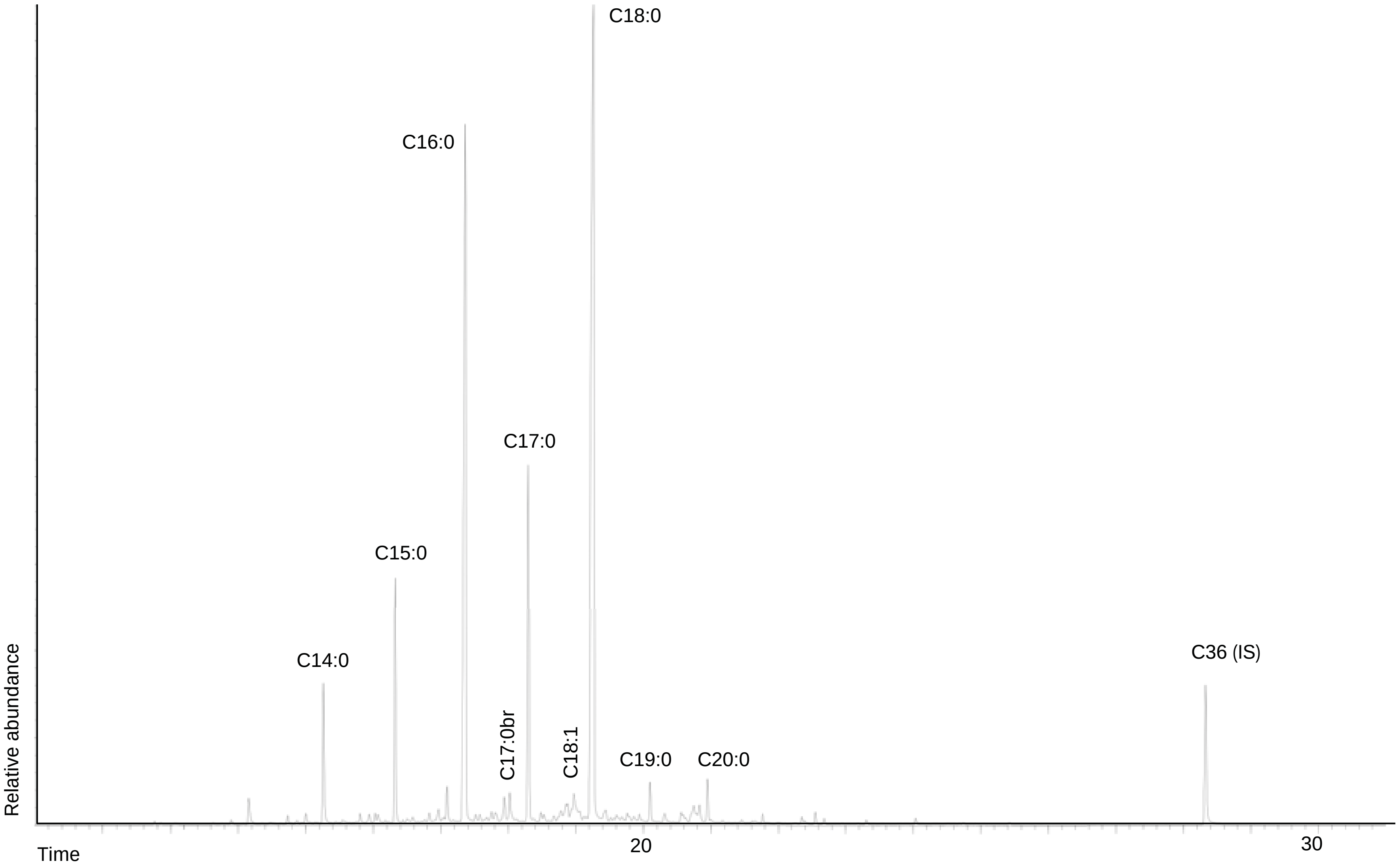
RA 8439


C36 (IS)
C16:0
C18:0
P
C18:1
C16:1
C15:0
Relative abundance
Time
20
30

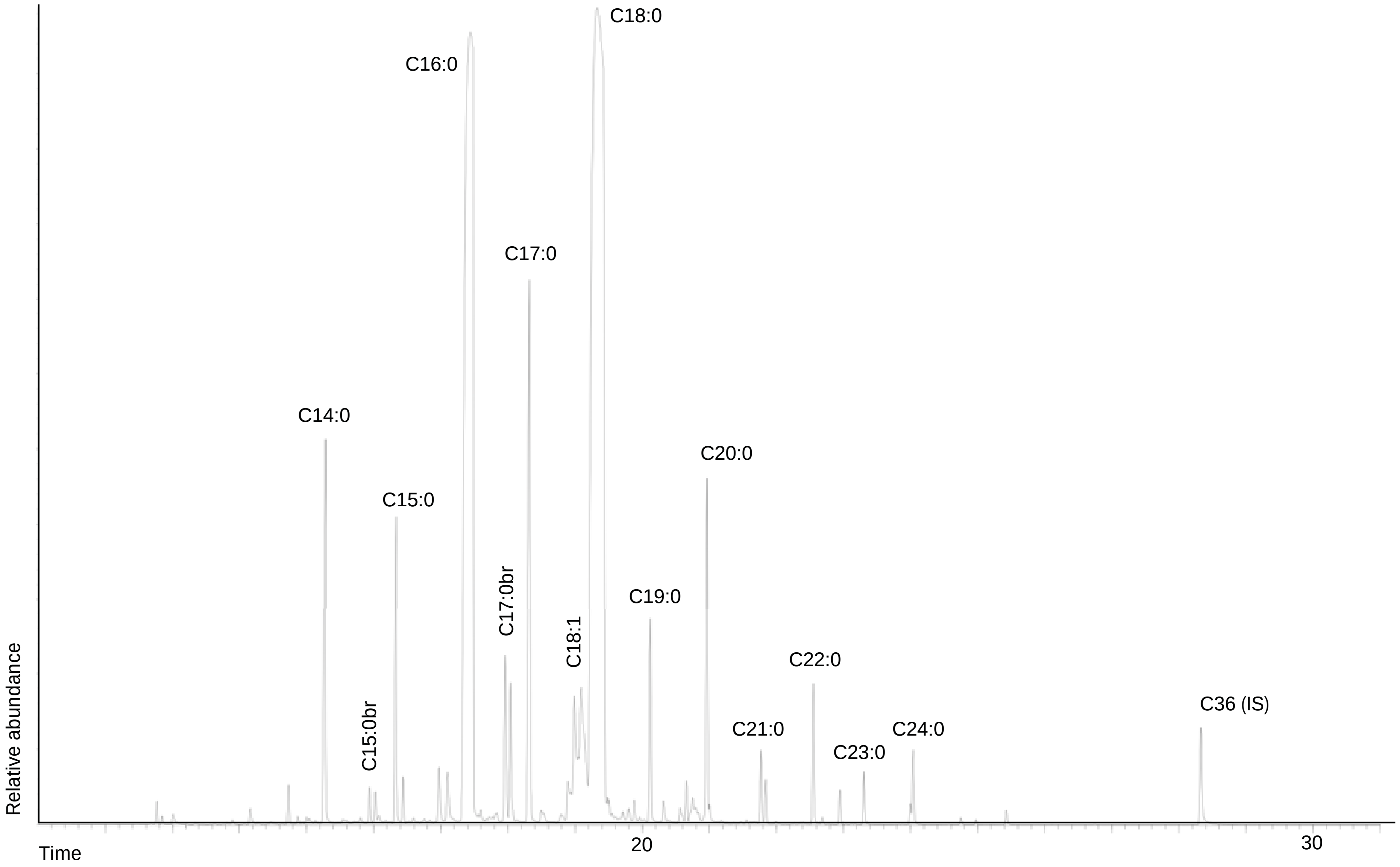
RA 8721


C18:0
C16:0
C17:0
C14:0
C15:0
C36 (IS)
C15:0br
C16:1
C17:0br
C18:1
C19:0
C20:0
C22:1
Relative abundance
Time
20
30

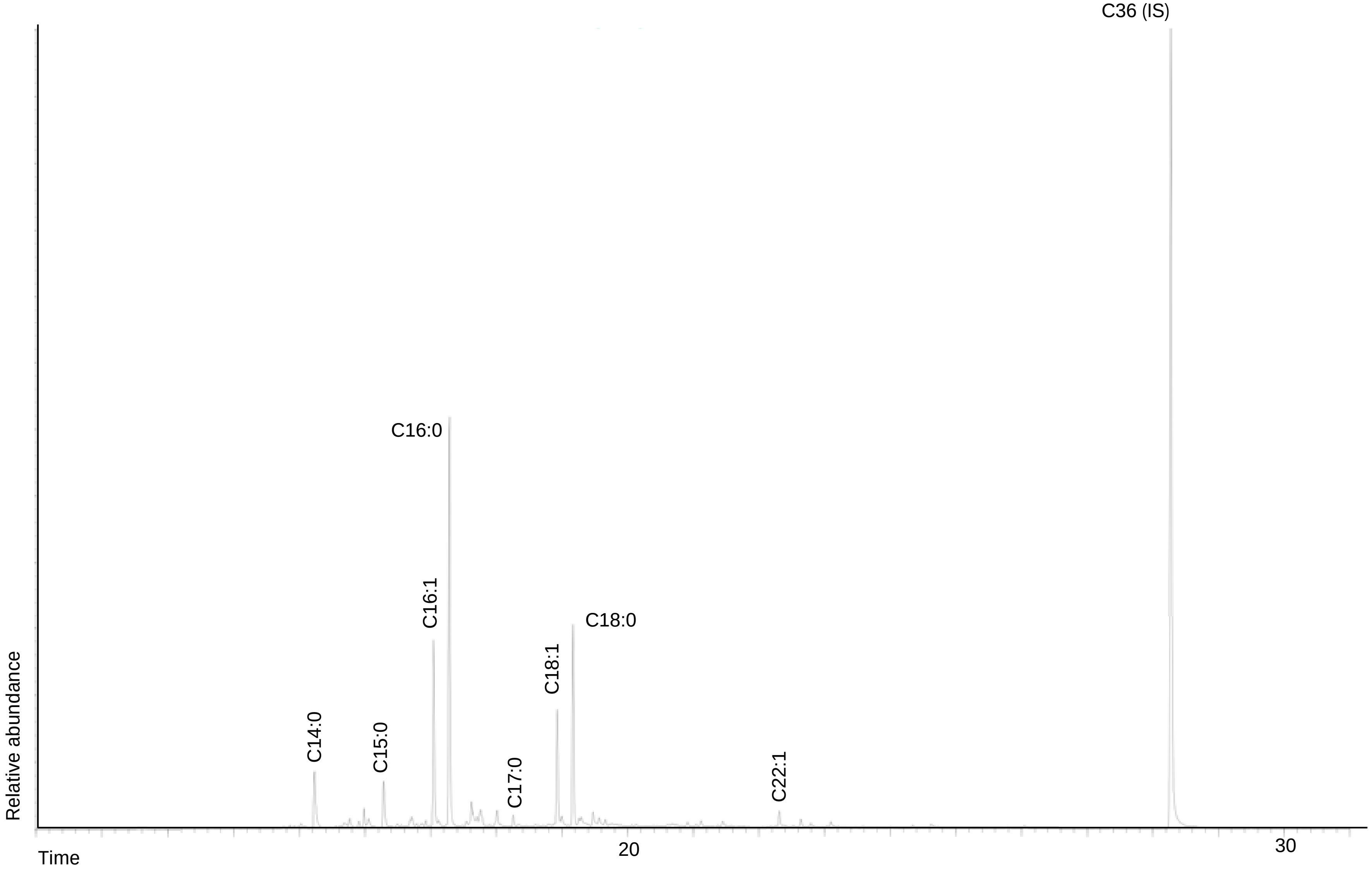
RA 8726


C18:0
C16:0
C17:0
C15:0
C14:0
C36 (IS)
C17:0br
C18:1
C19:0
C20:0
Relative abundance
Time
20
30

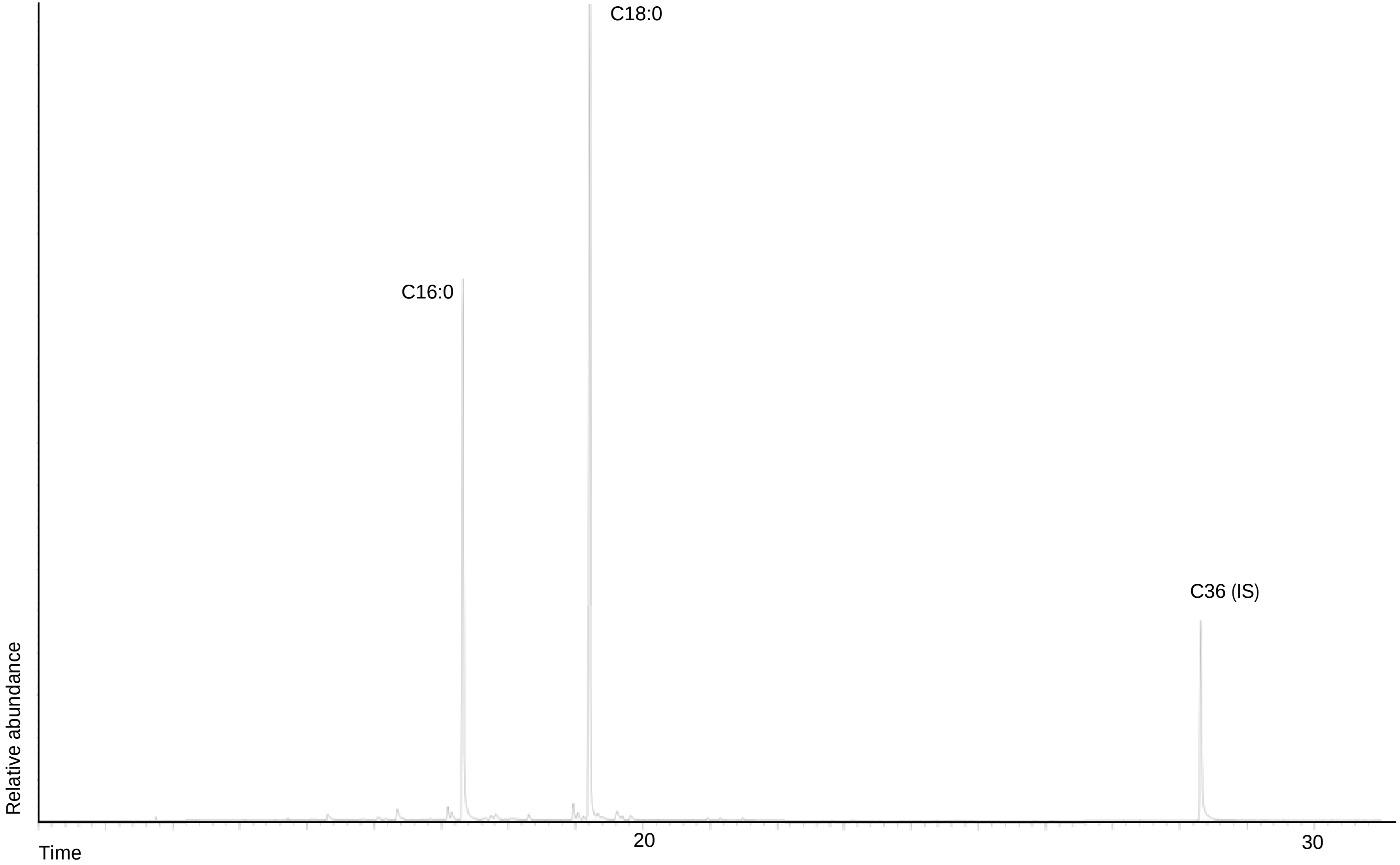
RA 8737


C18:0
C16:0
C17:0
C14:0
C20:0
C15:0
C17:0br
C18:1
C19:0
C22:0
C15:0br
C36 (IS)
C21:0
C24:0
C23:0
Relative abundance
Time
20
30

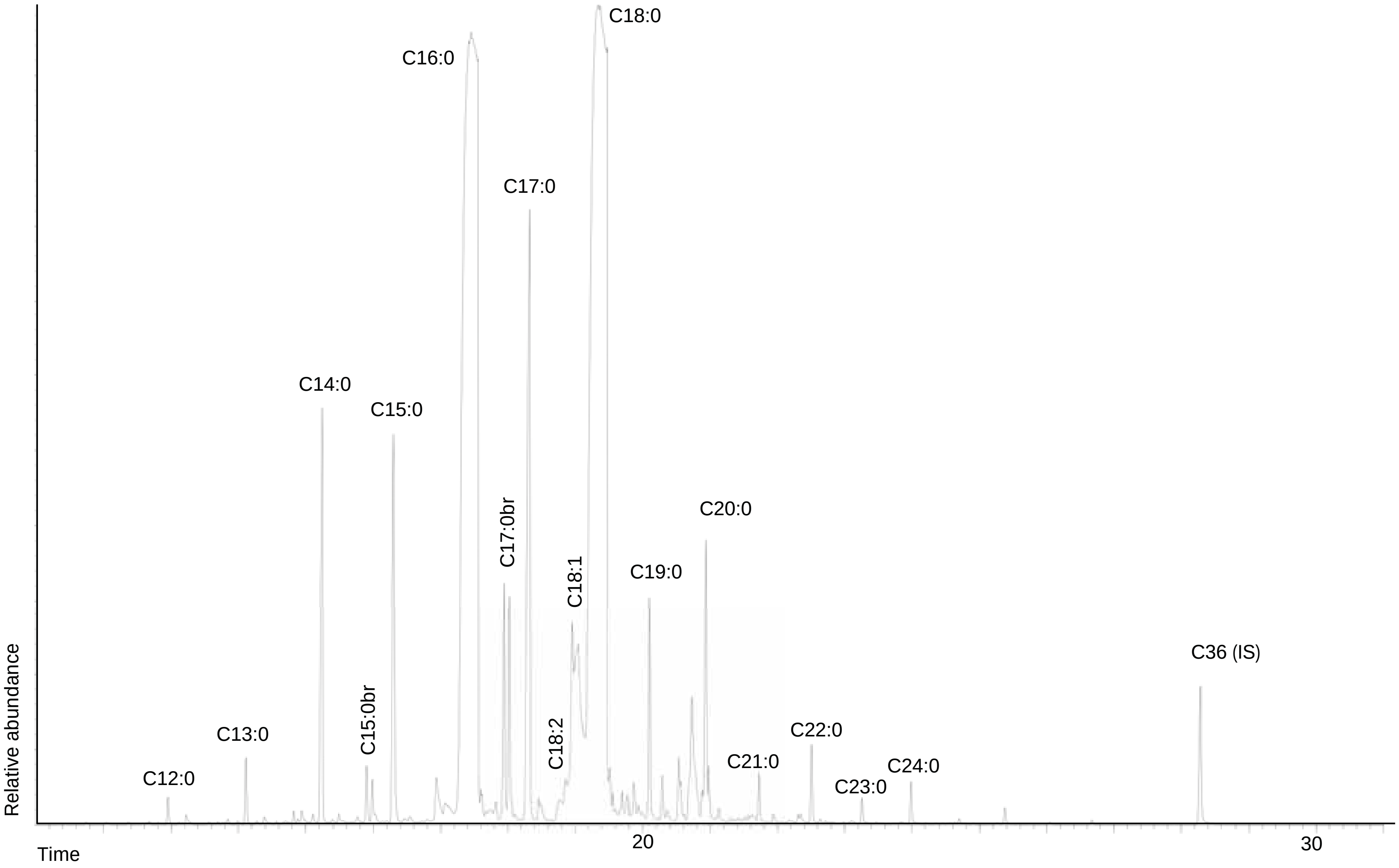
RA 8764


C36 (IS)
C16:0
C16:1
C18:0
C18:1
C14:0
C15:0
C17:0
C22:1
Relative abundance
Time
20
30

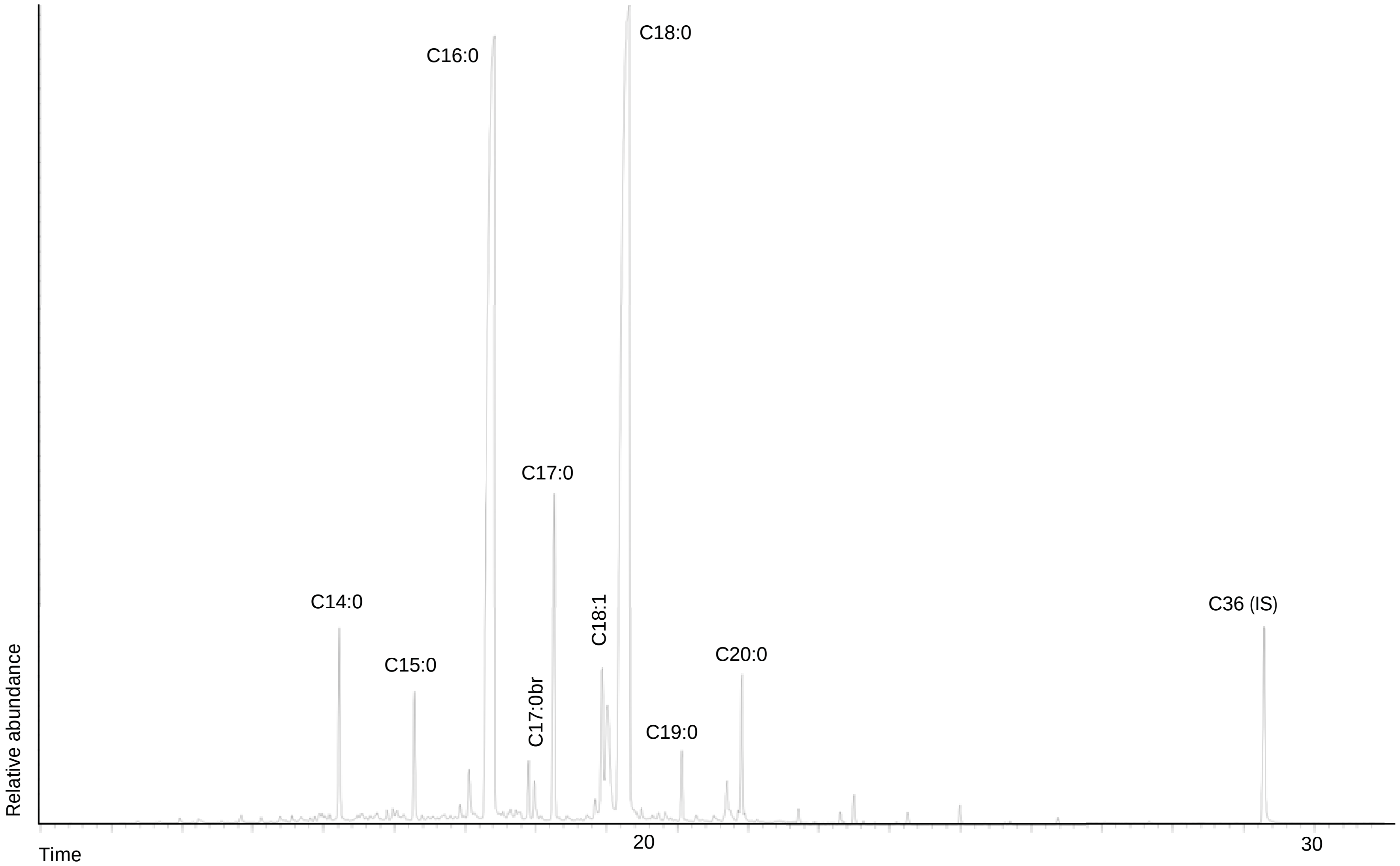
RA 8842



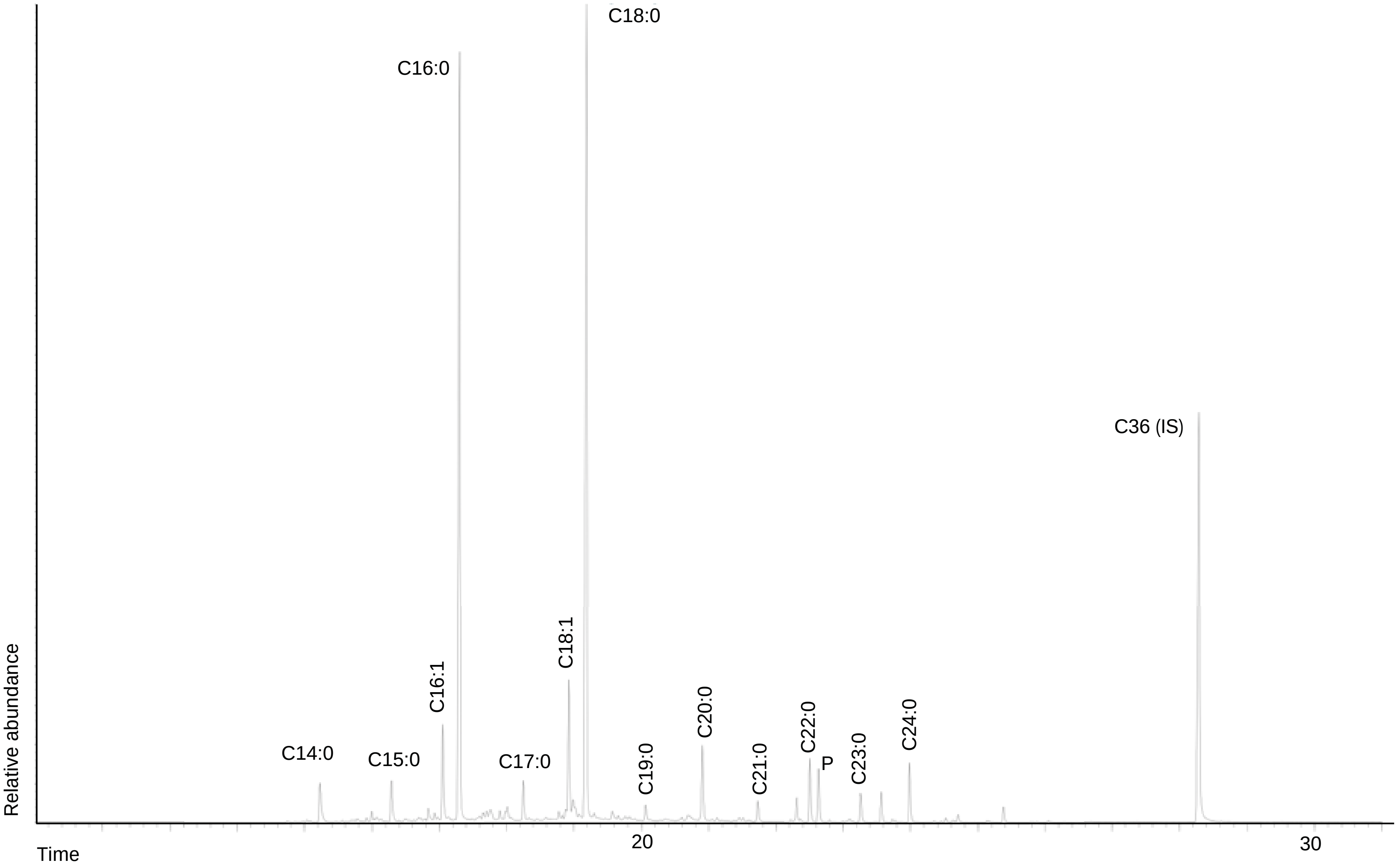
RA 8995


C18:0
C16:0
C17:0
C14:0
C15:0
C20:0
C17:0br
C18:1
C19:0
C36 (IS)
C13:0
C15:0br
C18:2
C22:0
C12:0
C21:0
C24:0
C23:0
Relative abundance
Time
20
30

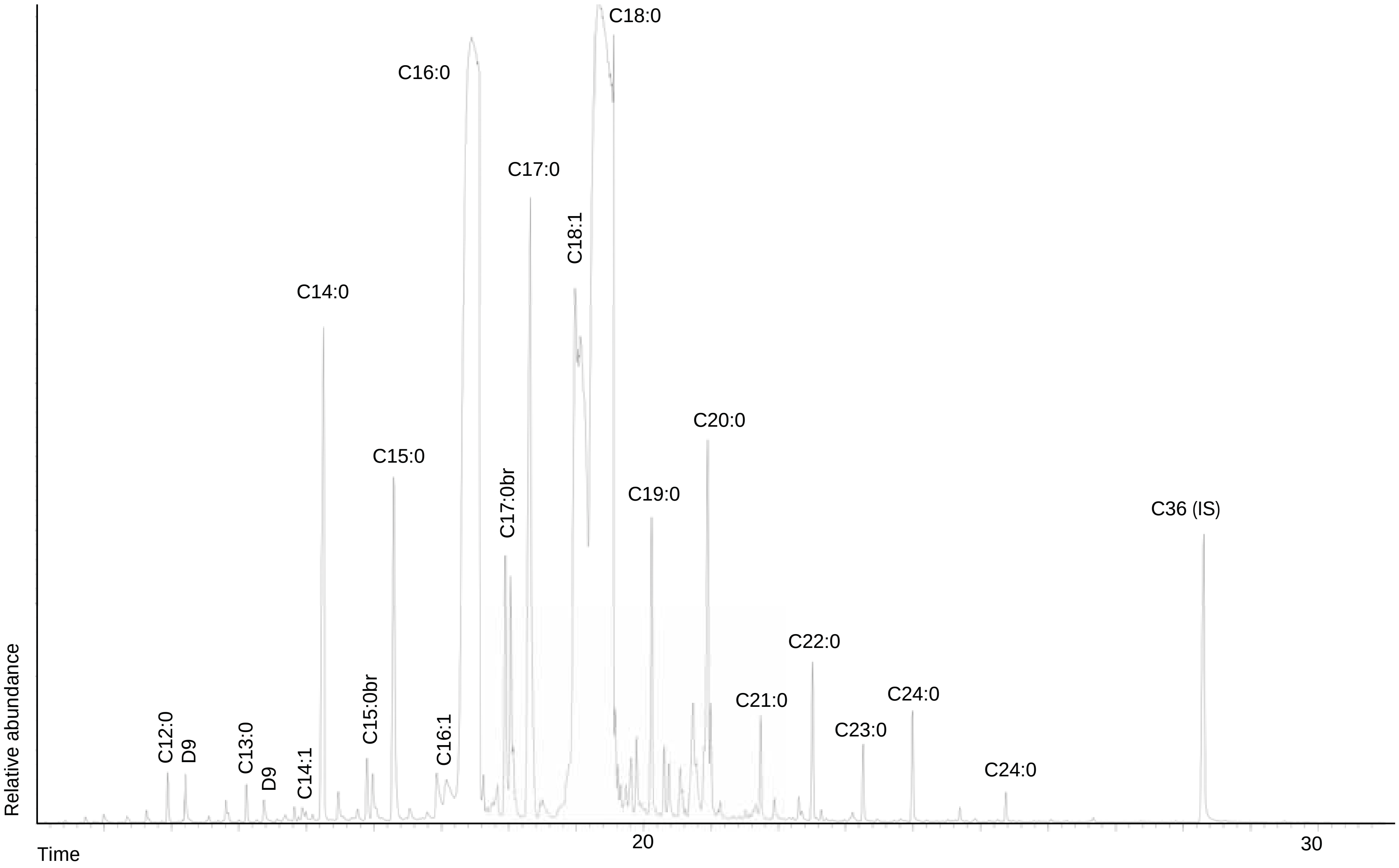
RA 9064


C16:0
C18:0
C17:0
C14:0
C15:0
C17:0br
C18:1
C19:0
C20:0
C36 (IS)
Relative abundance
Time
20
30

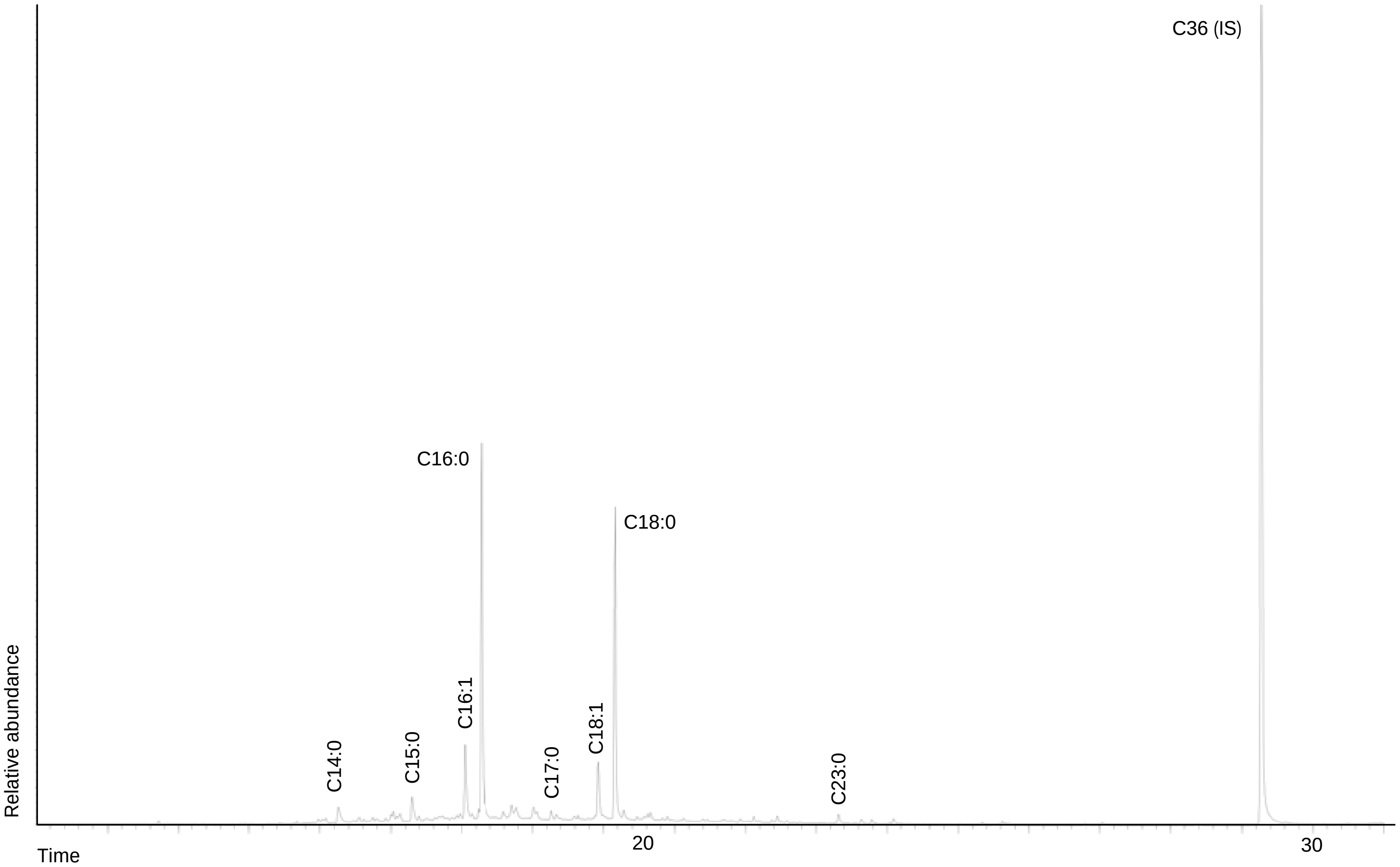
RA 9065


C18:0
C16:0
C36 (IS)
C18:1
C16:1
C20:0
C22:0
C24:0
C14:0
C15:0
C17:0
C19:0
C21:0
P
C23:0
Relative abundance
Time
20
30

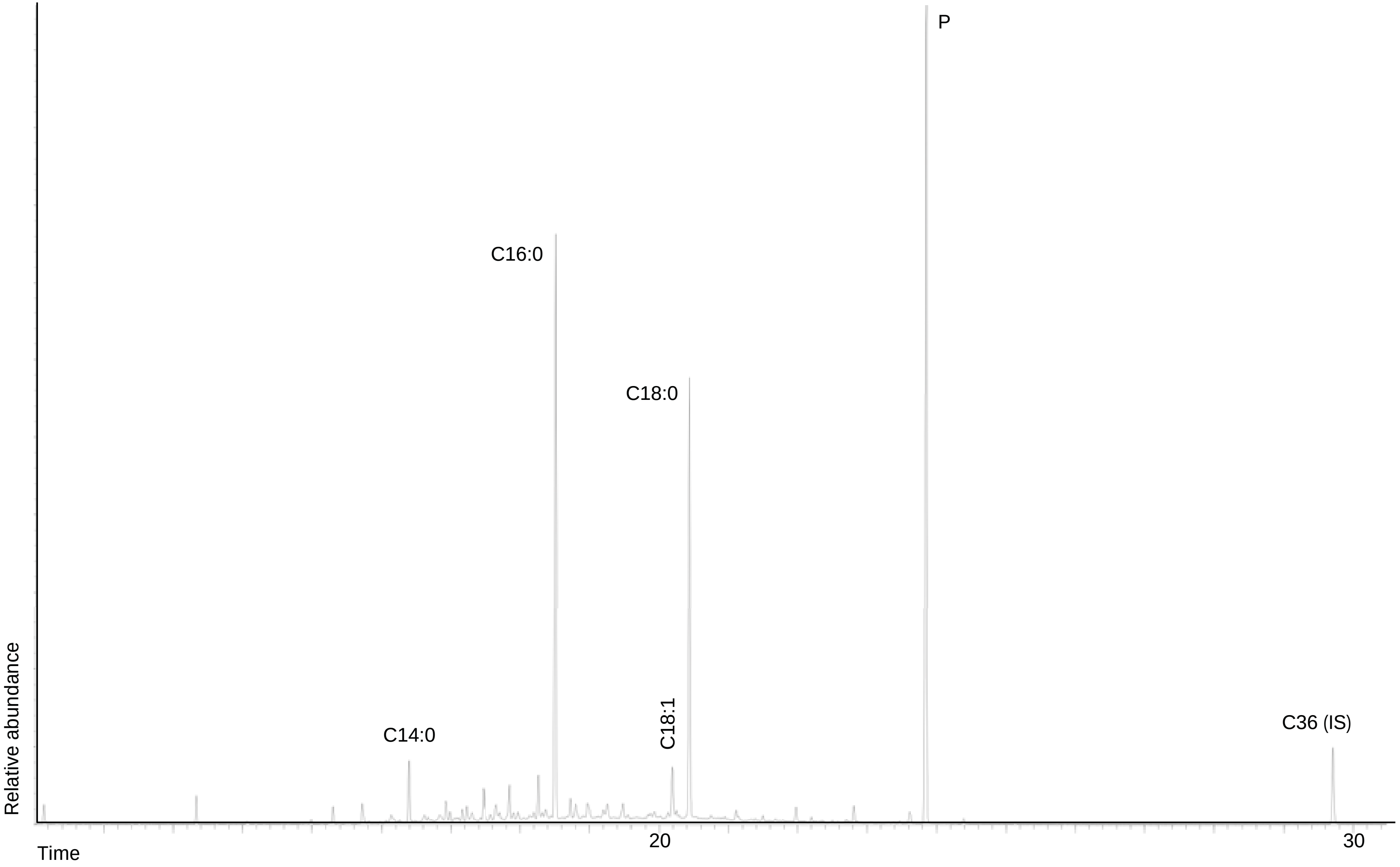
RA 9464


Relative abundance
C12:0
D9
C13:0
D9
C14:1
C14:0
C15:0br
C15:0
C16:1
C16:0
C17:0br
C17:0
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C24:0
C36 (IS)
Time
20
30

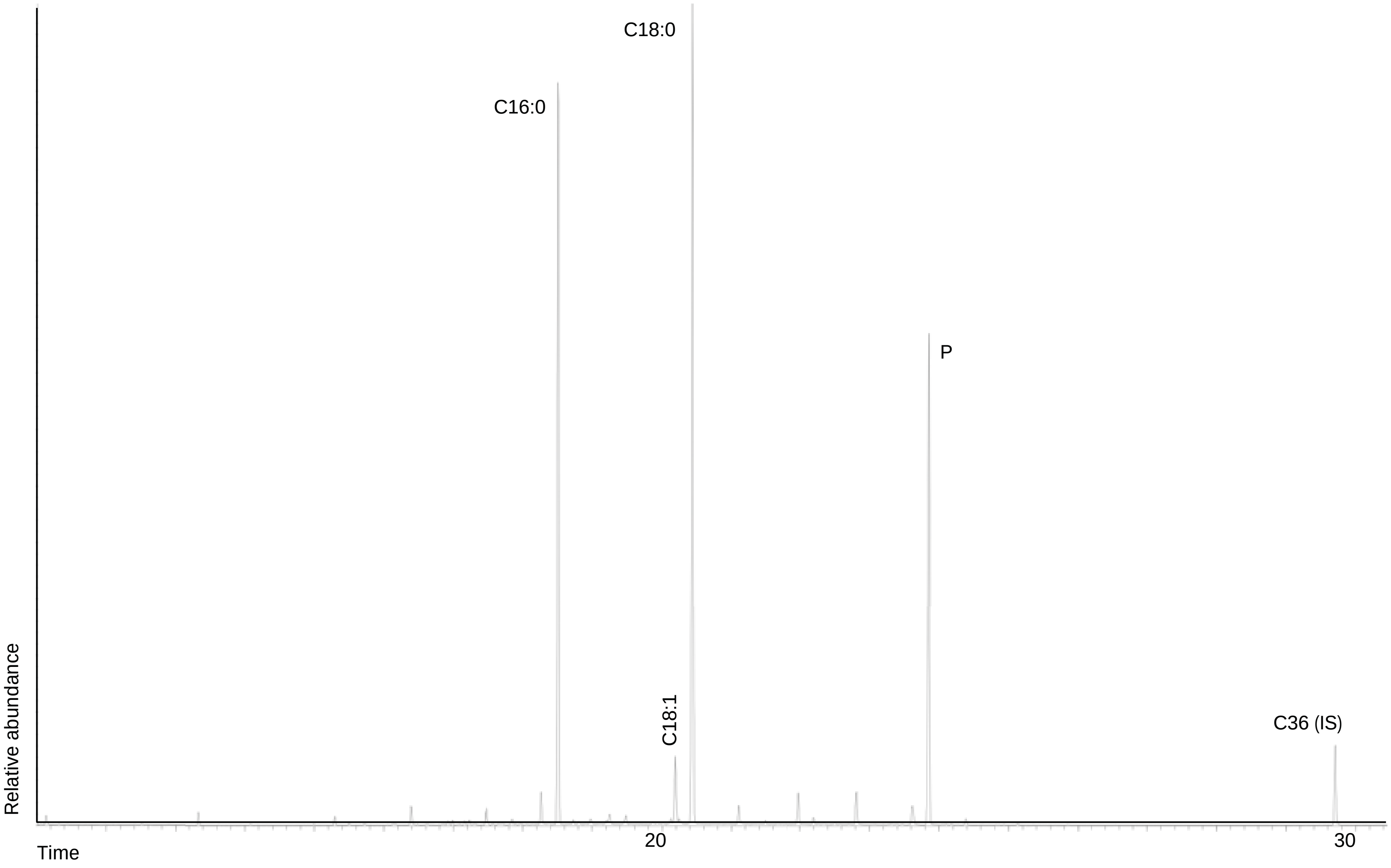
RA 11960


C36 (IS)
C16:0
C18:0
C14:0
C15:0
C16:1
C17:0
C18:1
C23:0
Relative abundance
Time
20
30

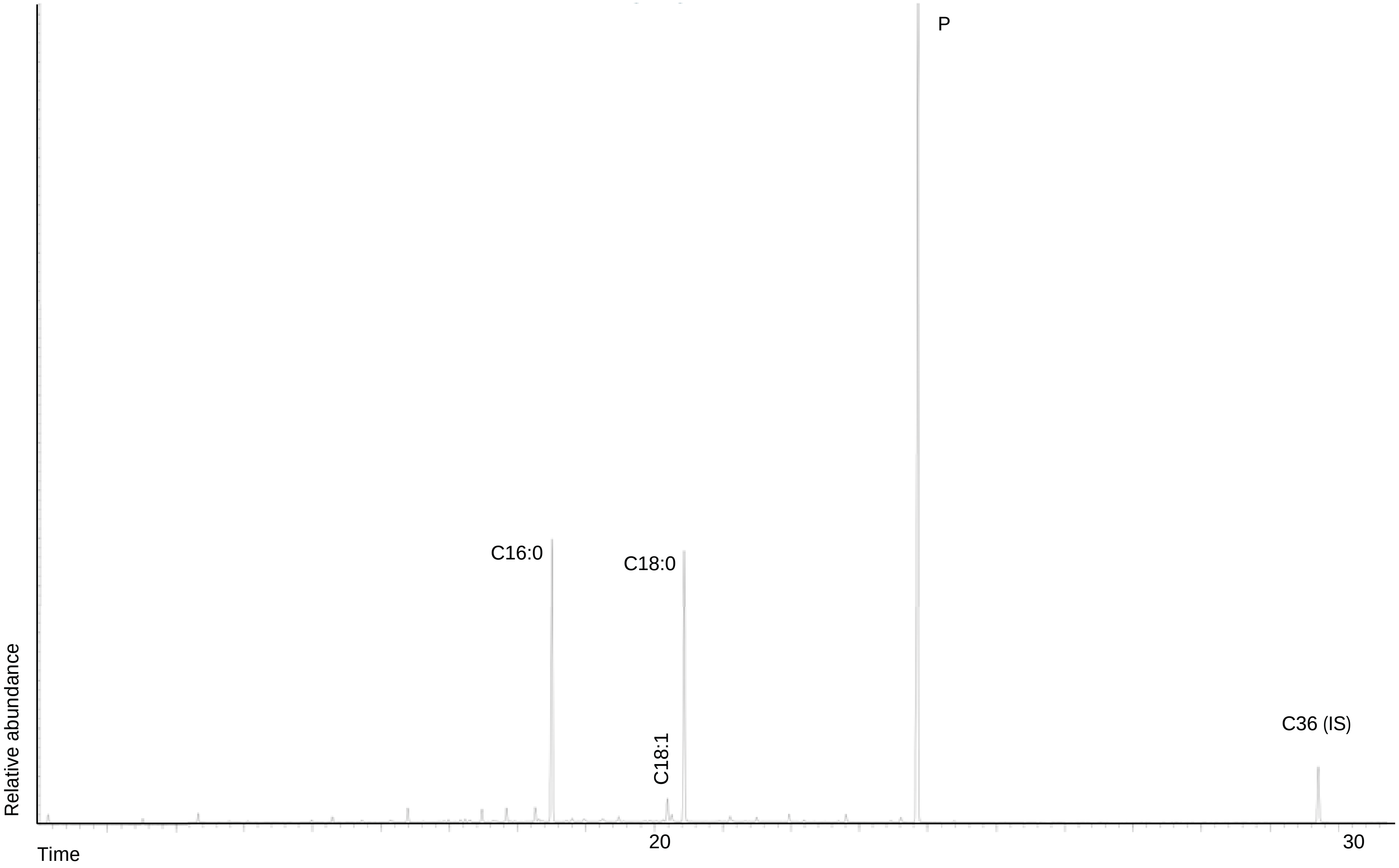
RA 8090



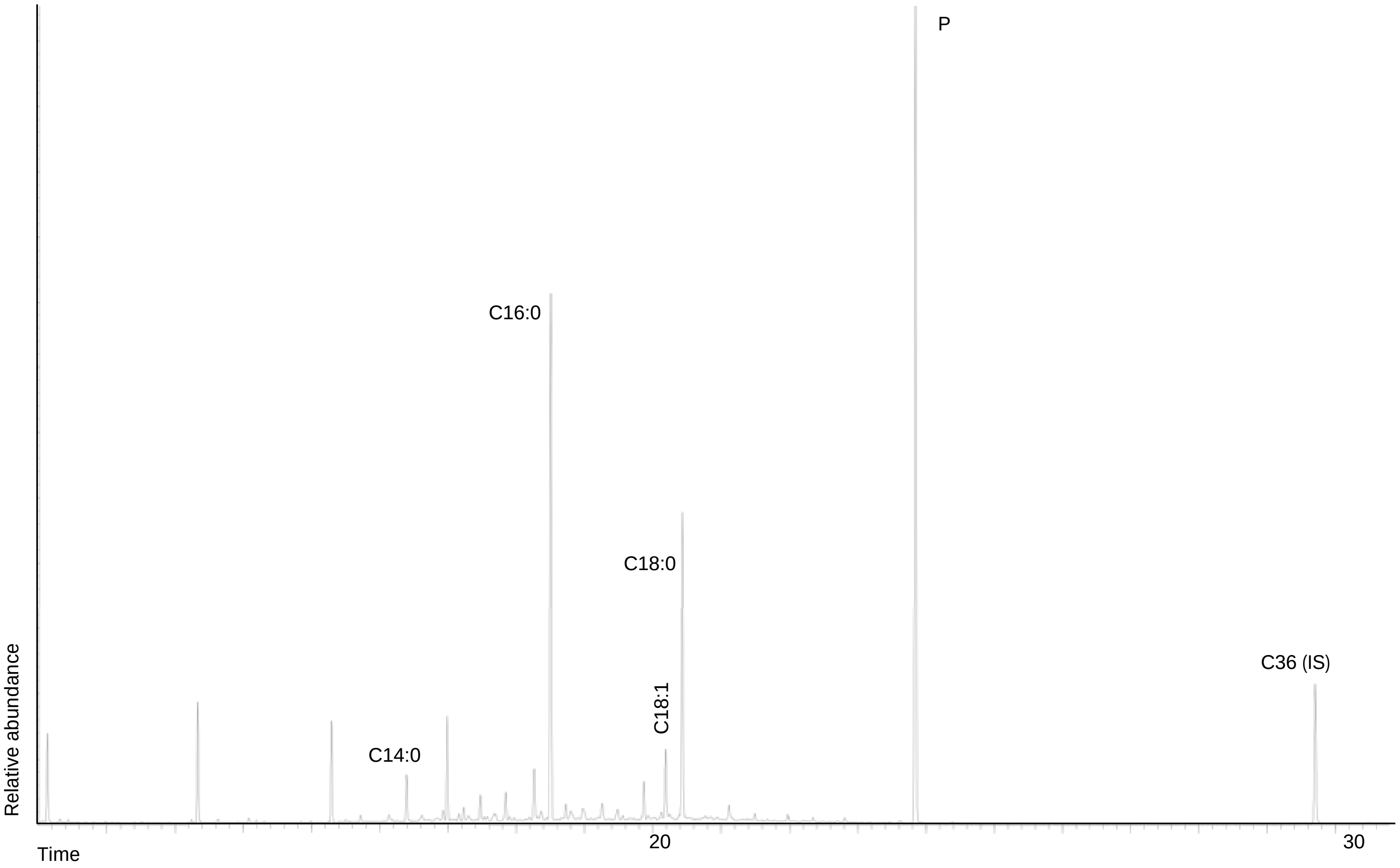
RA 8656



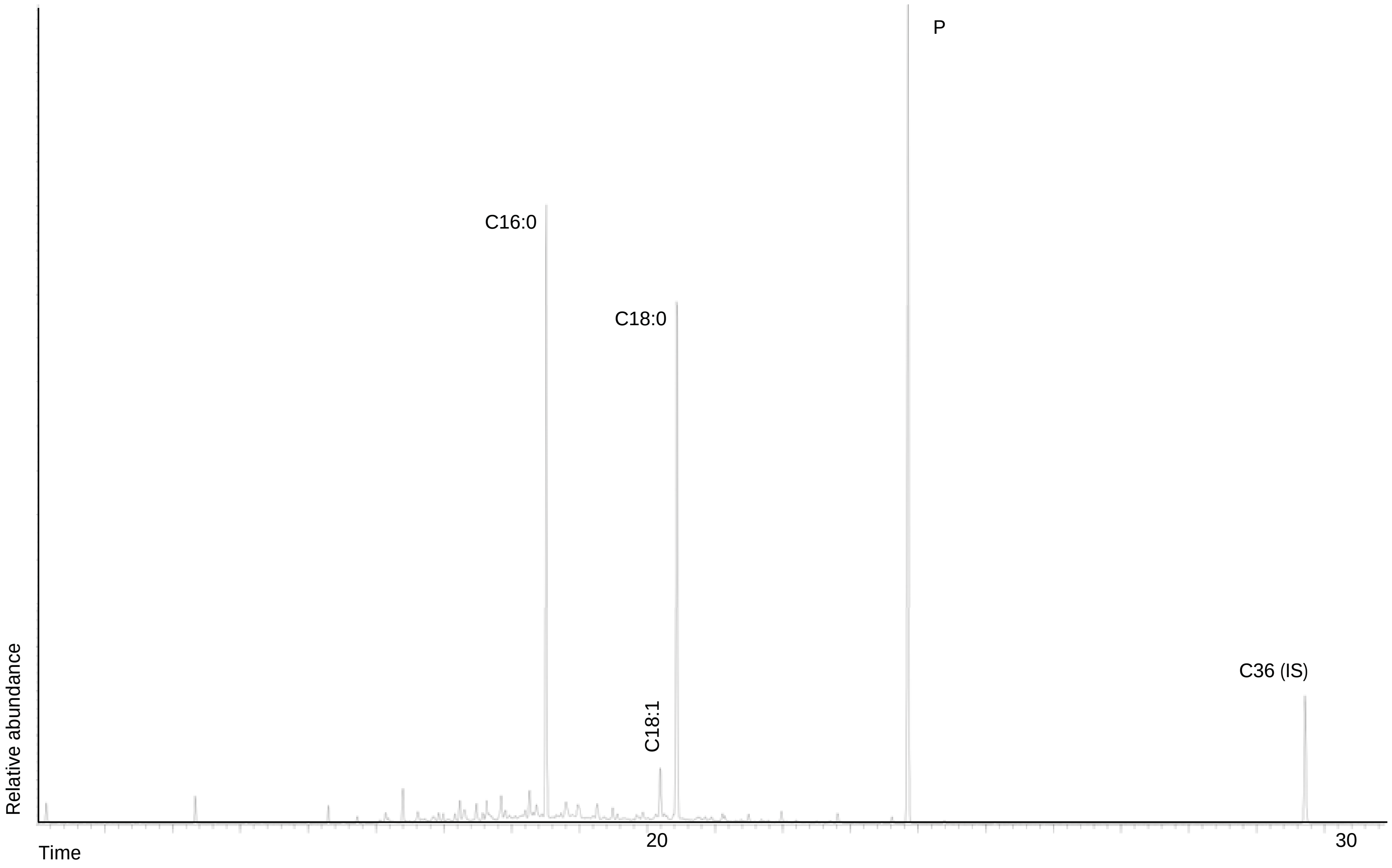
RA 8890



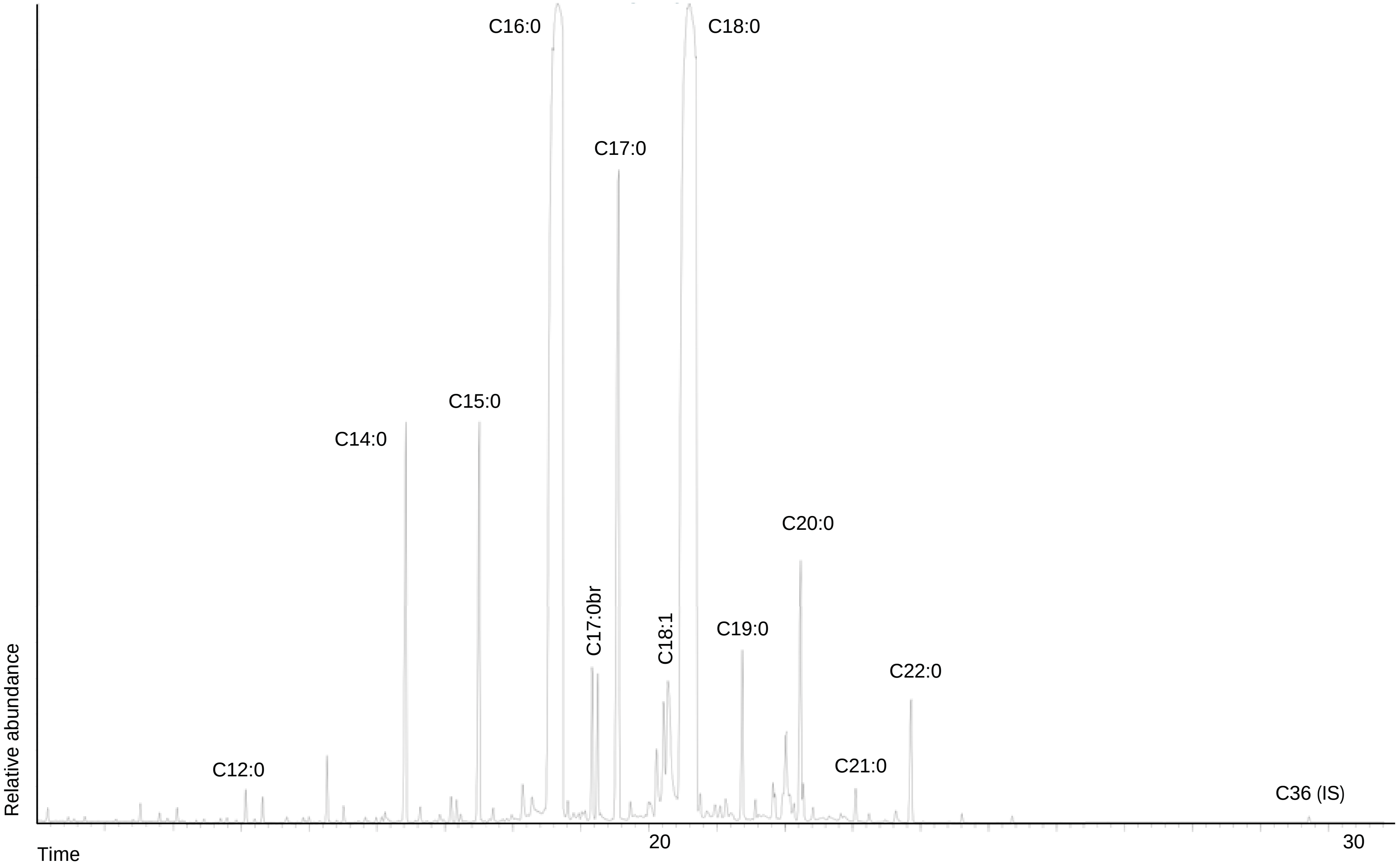
RA 8900



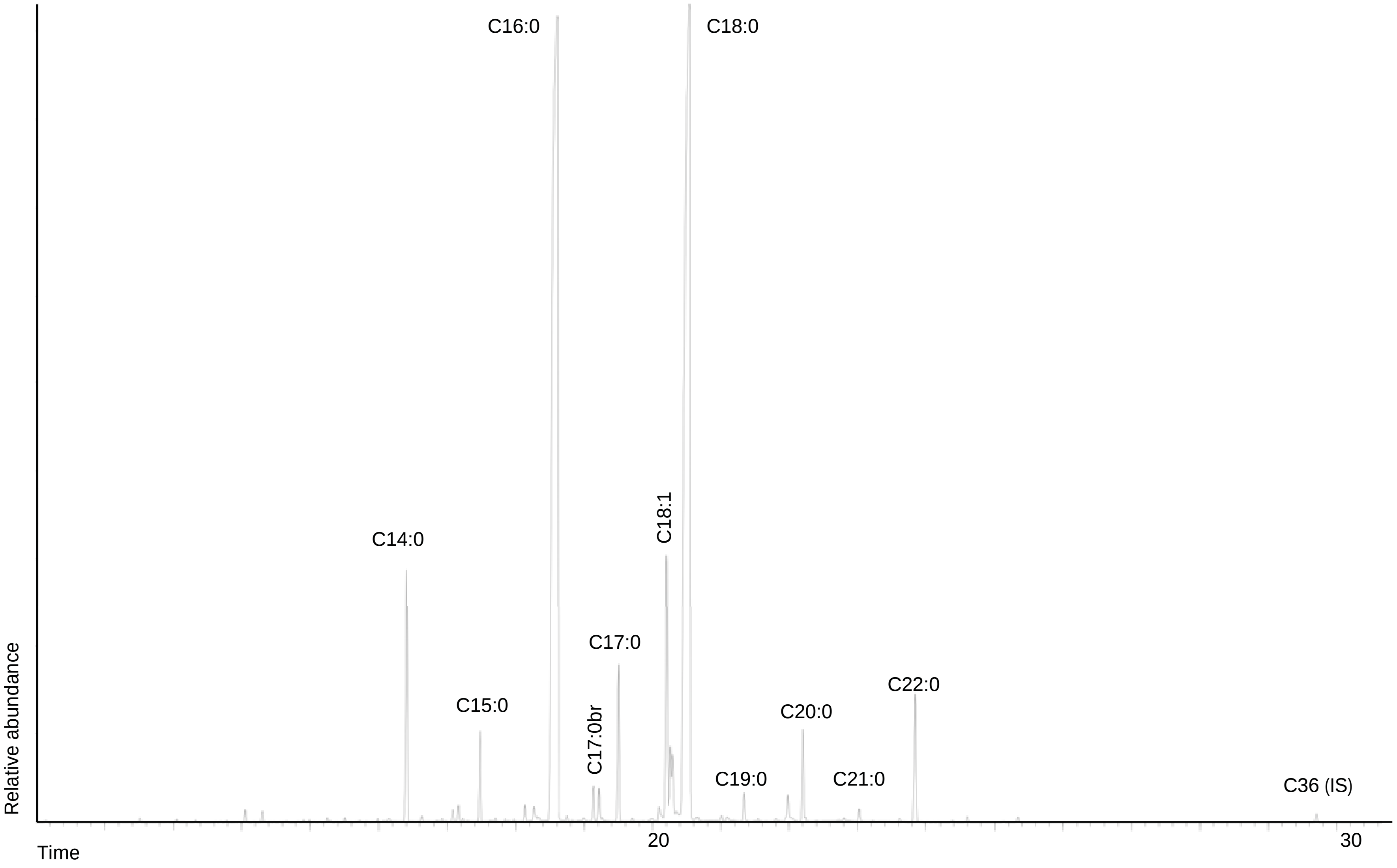
RA 9086



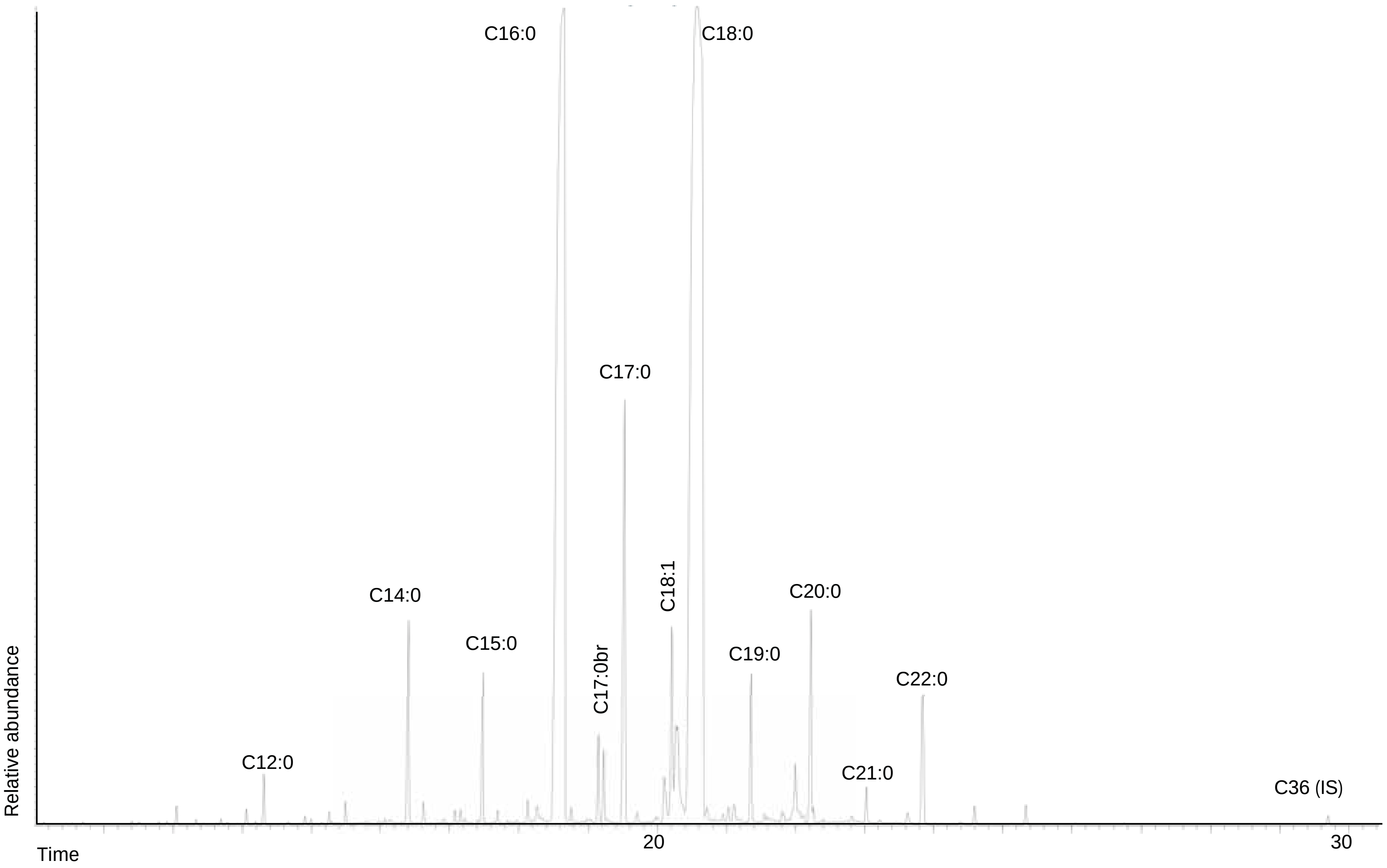
RA 9092


C16:0
C18:0
C17:0
C15:0
C14:0
C20:0
C17:0br
C18:1
C19:0
C22:0
C12:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

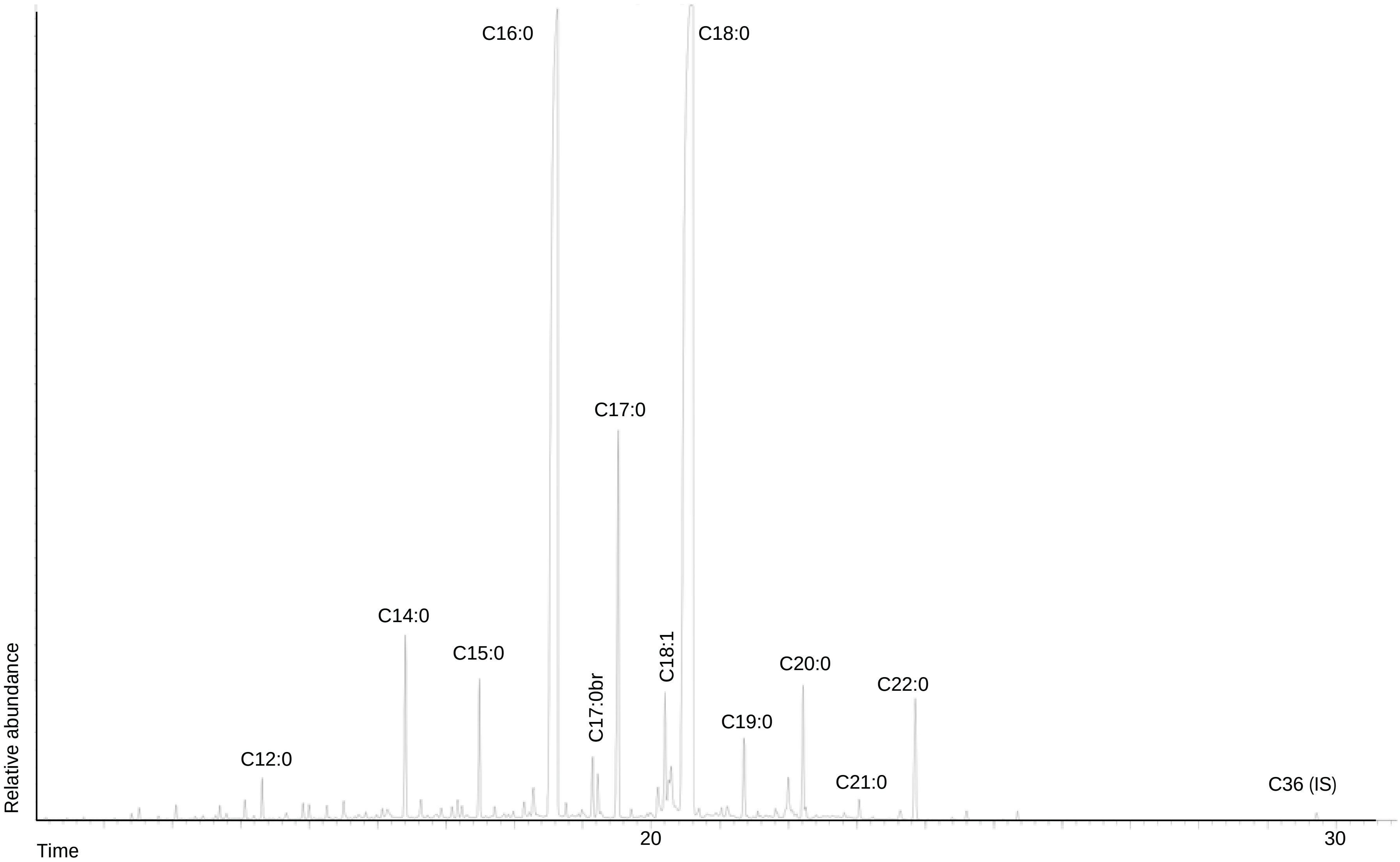
RA 9189


C16:0
C18:0
C14:0
C18:1
C17:0
C15:0
C17:0br
C22:0
C20:0
C19:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

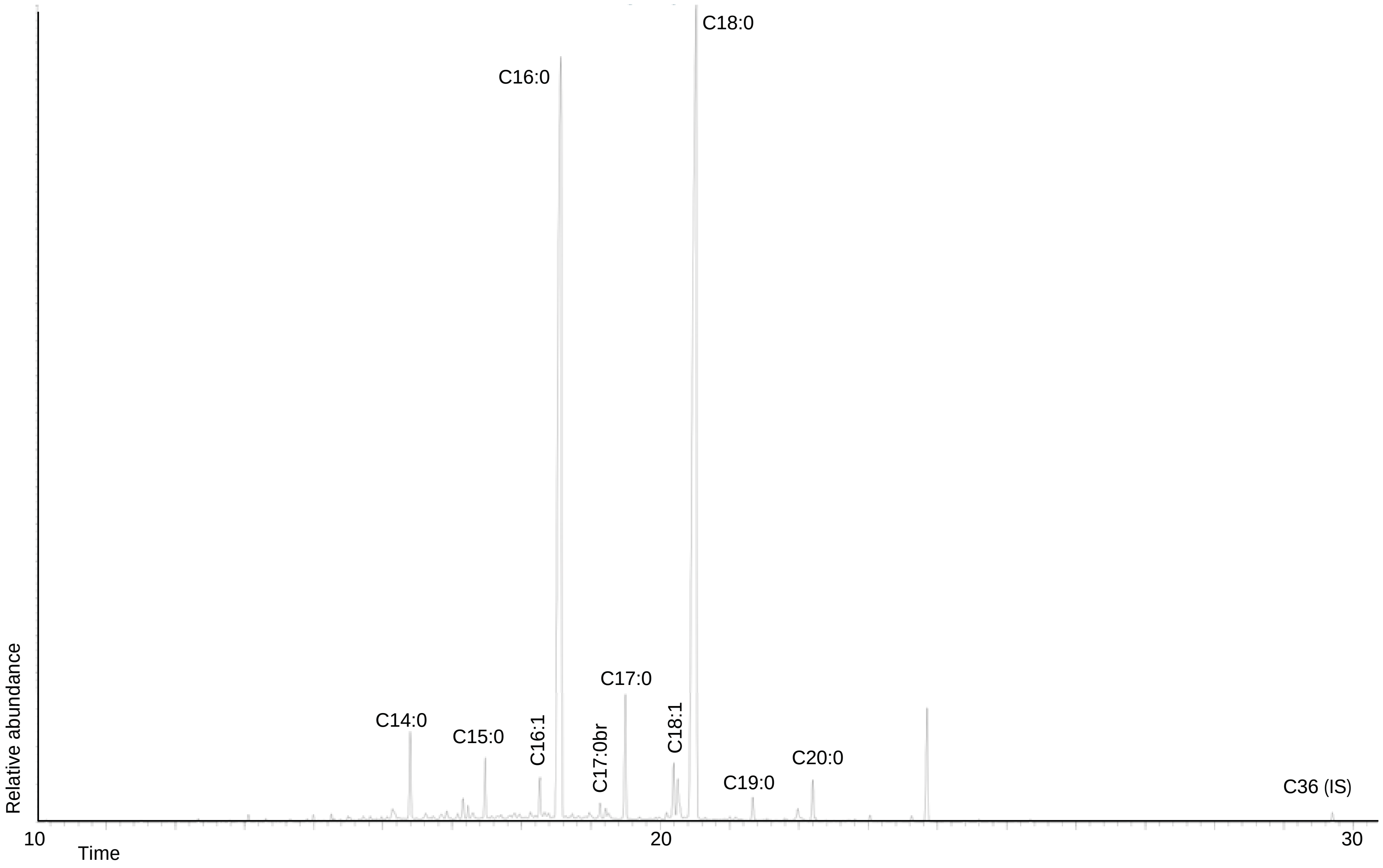
RA 9296


C16:0
C18:0
C17:0
C14:0
C18:1
C20:0
C15:0
C17:0br
C19:0
C22:0
C12:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

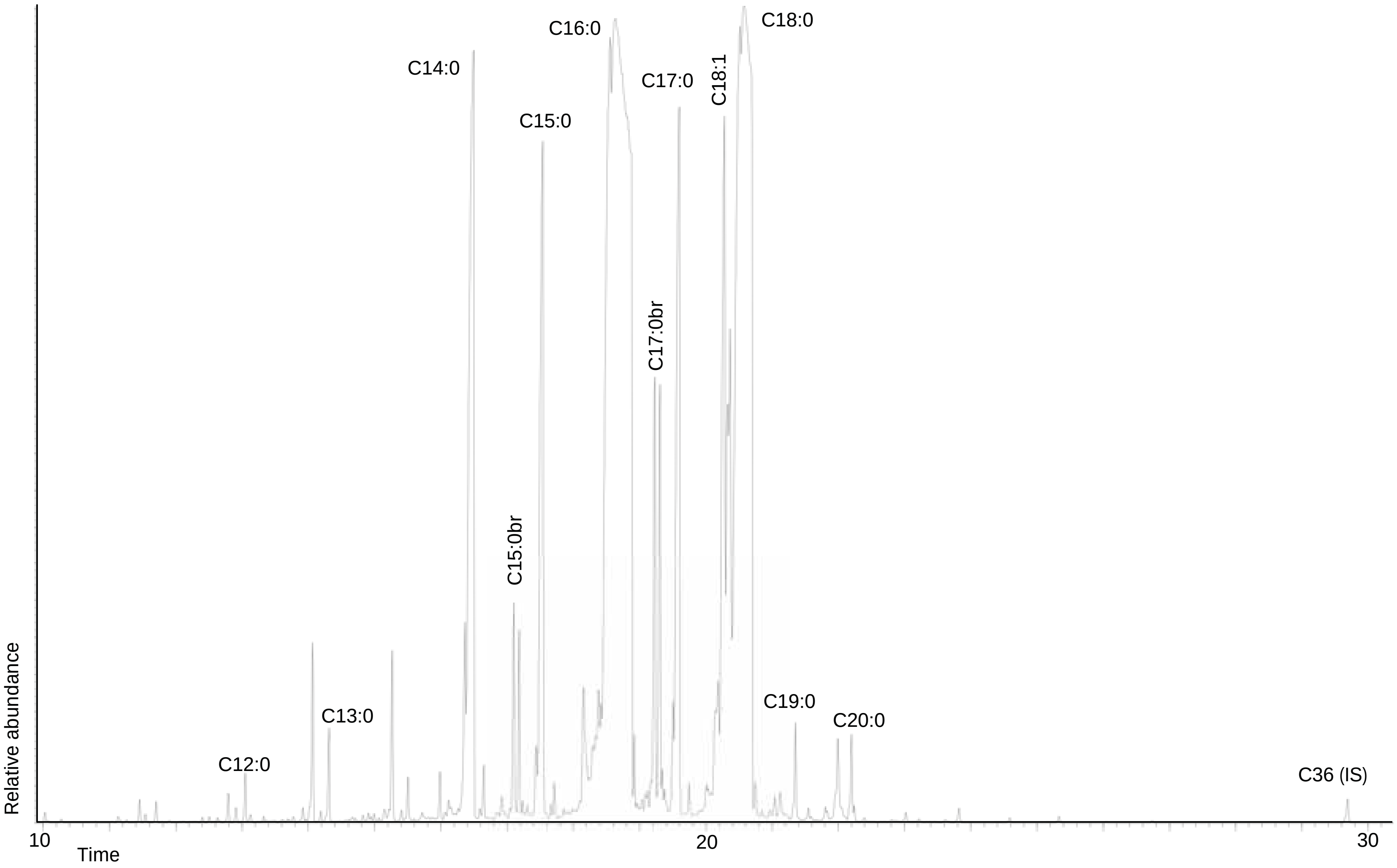
RA 9343


C16:0
C18:0
C17:0
C14:0
C15:0
C17:0br
C18:1
C20:0
C22:0
C19:0
C12:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

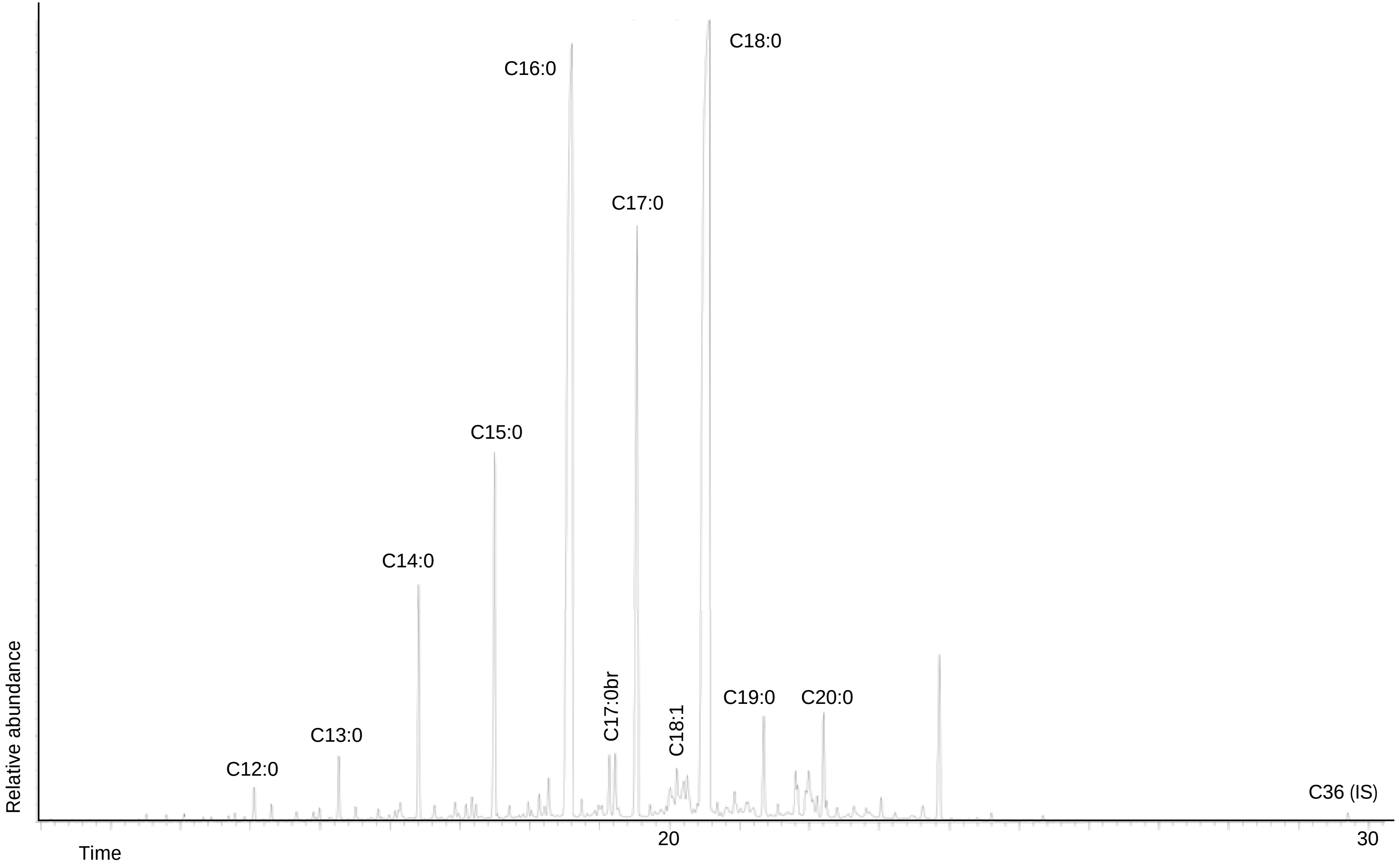
RA 9359


C18:0
C16:0
C17:0
C14:0
C15:0
C16:1
C17:0br
C18:1
C20:0
C19:0
C36 (IS)
Relative abundance
10
20
30
Time

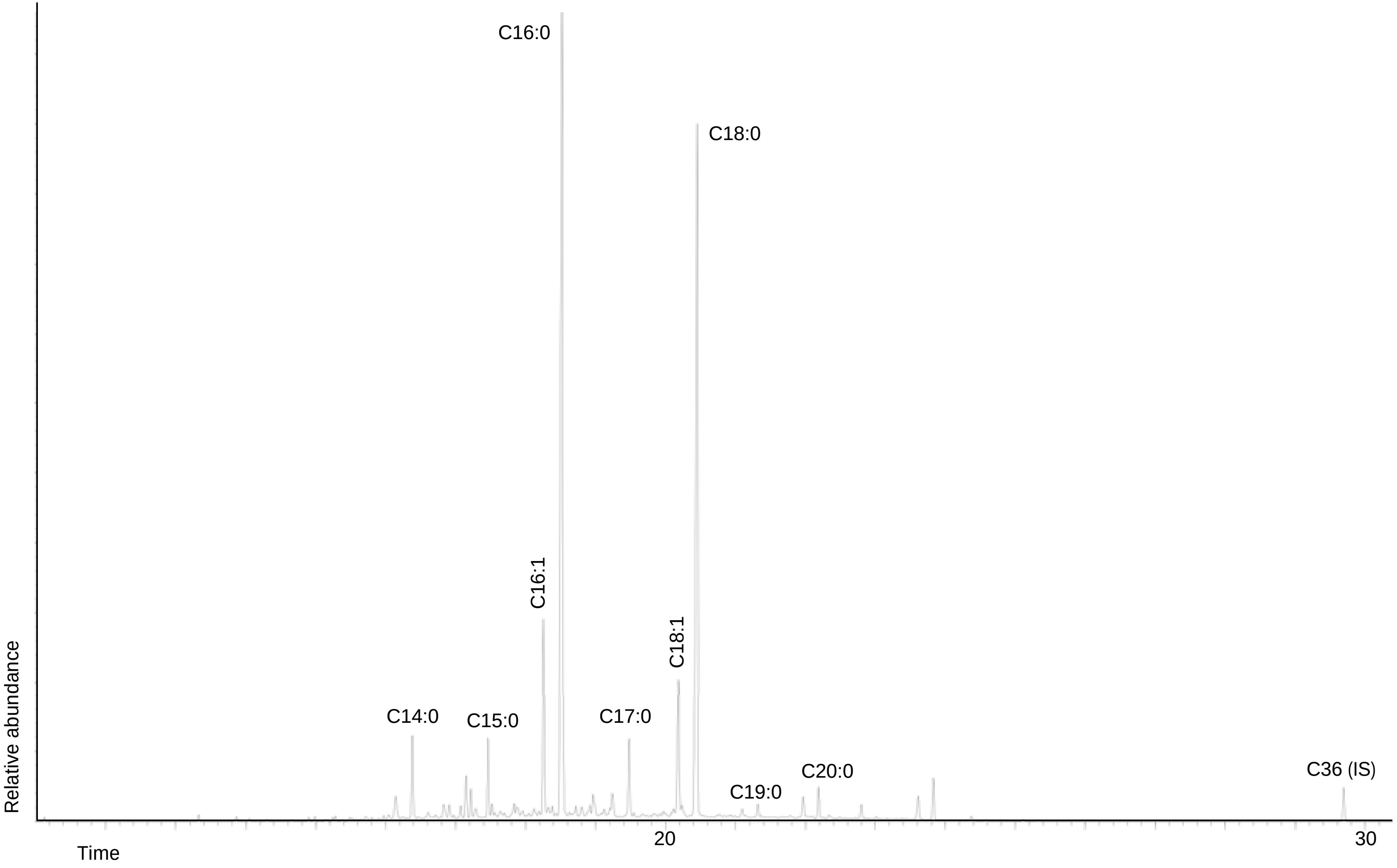
RA 9409


C14:0
C16:0
C18:0
C17:0
C18:1
C15:0
C17:0br
C15:0br
C12:0
C13:0
C19:0
C20:0
C36 (IS)
Relative abundance
10
20
30
Time

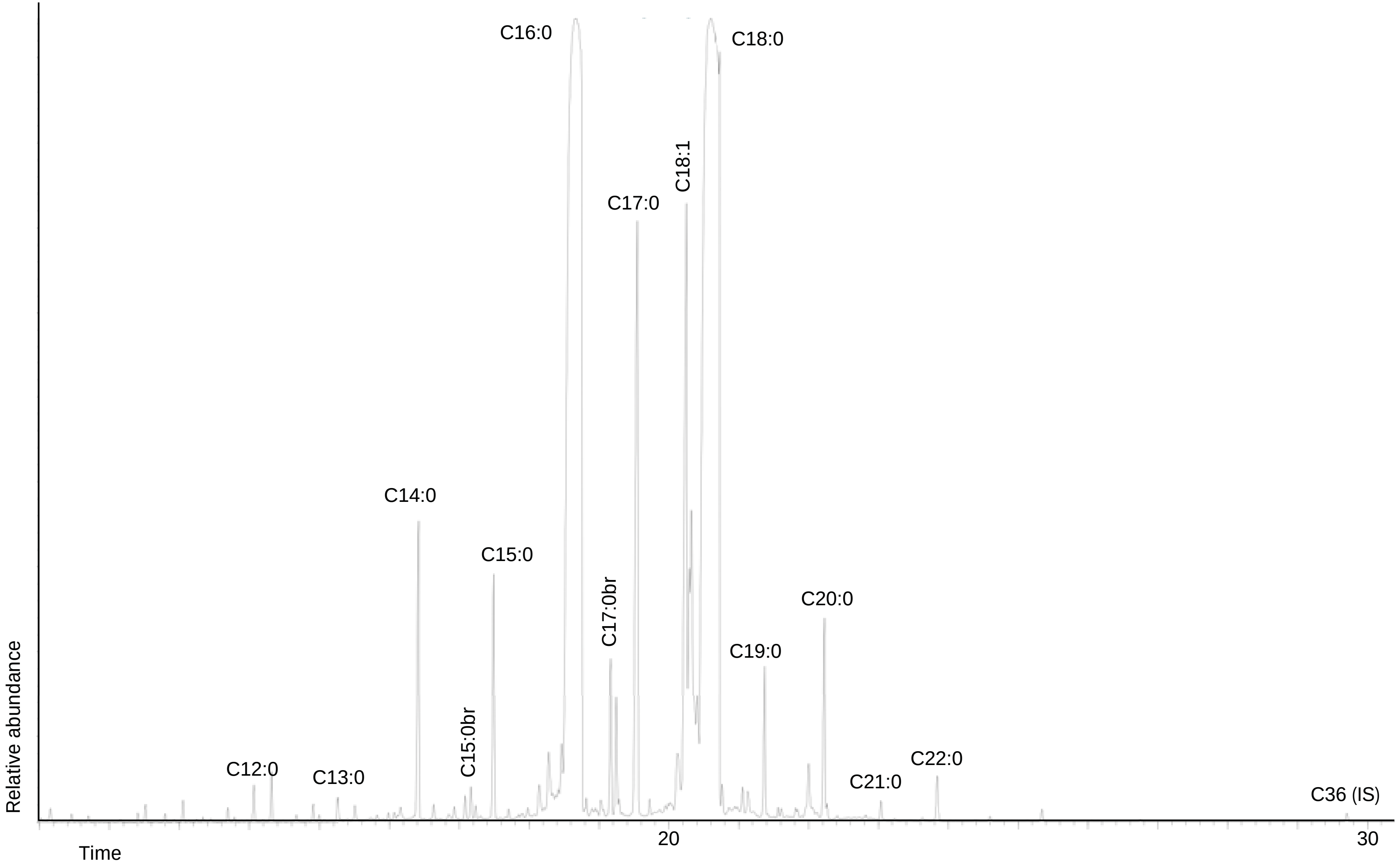
RA 9470


C12:0
C13:0
C14:0
C15:0
C16:0
C17:0br
C17:0
C18:1
C18:0
C19:0
C20:0
C36 (IS)
Relative abundance
Time
20
30

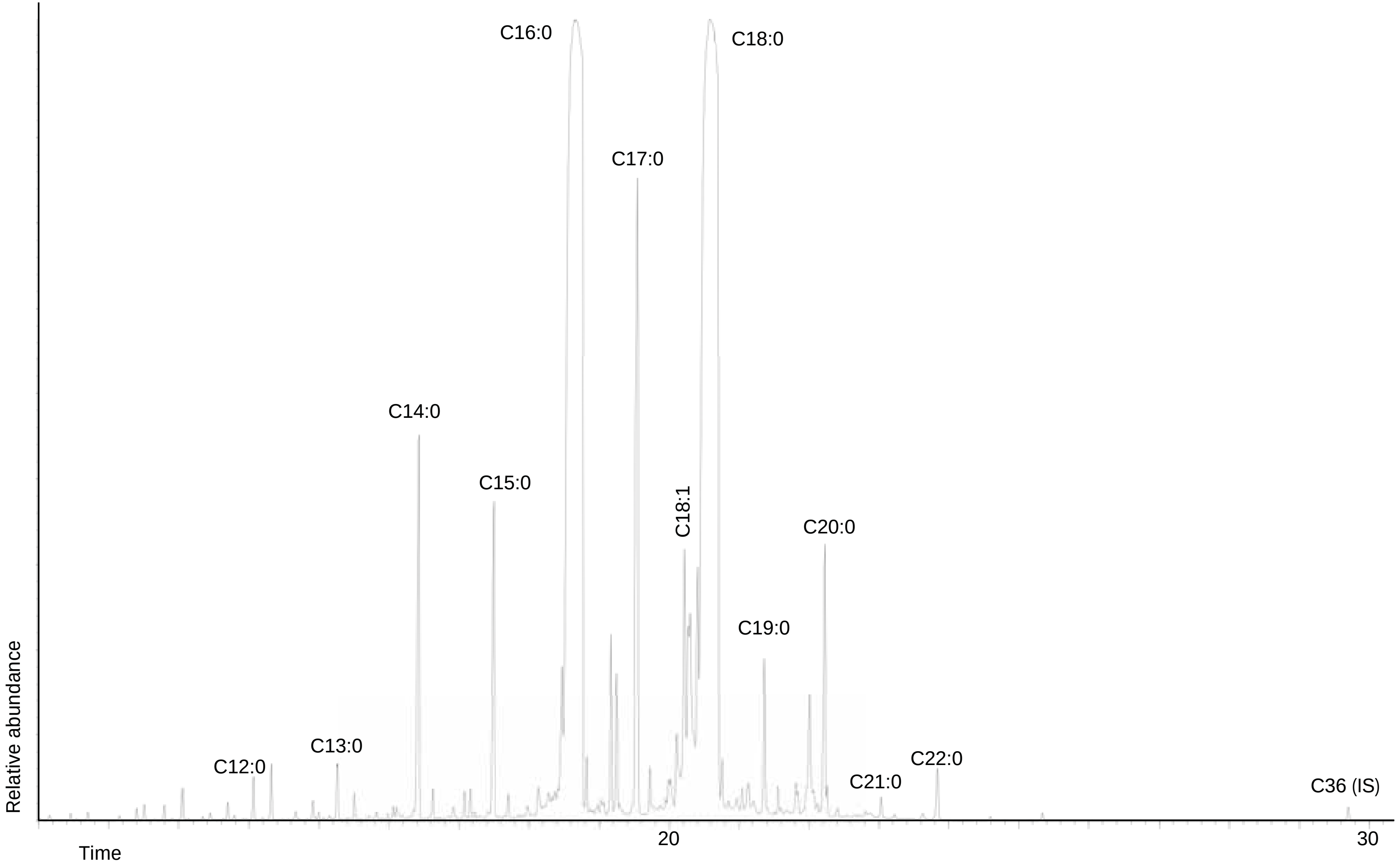
RA 9565


C16:0
C18:0
C16:1
C18:1
C14:0
C15:0
C17:0
C19:0
C20:0
C36 (IS)
Relative abundance
Time
20
30

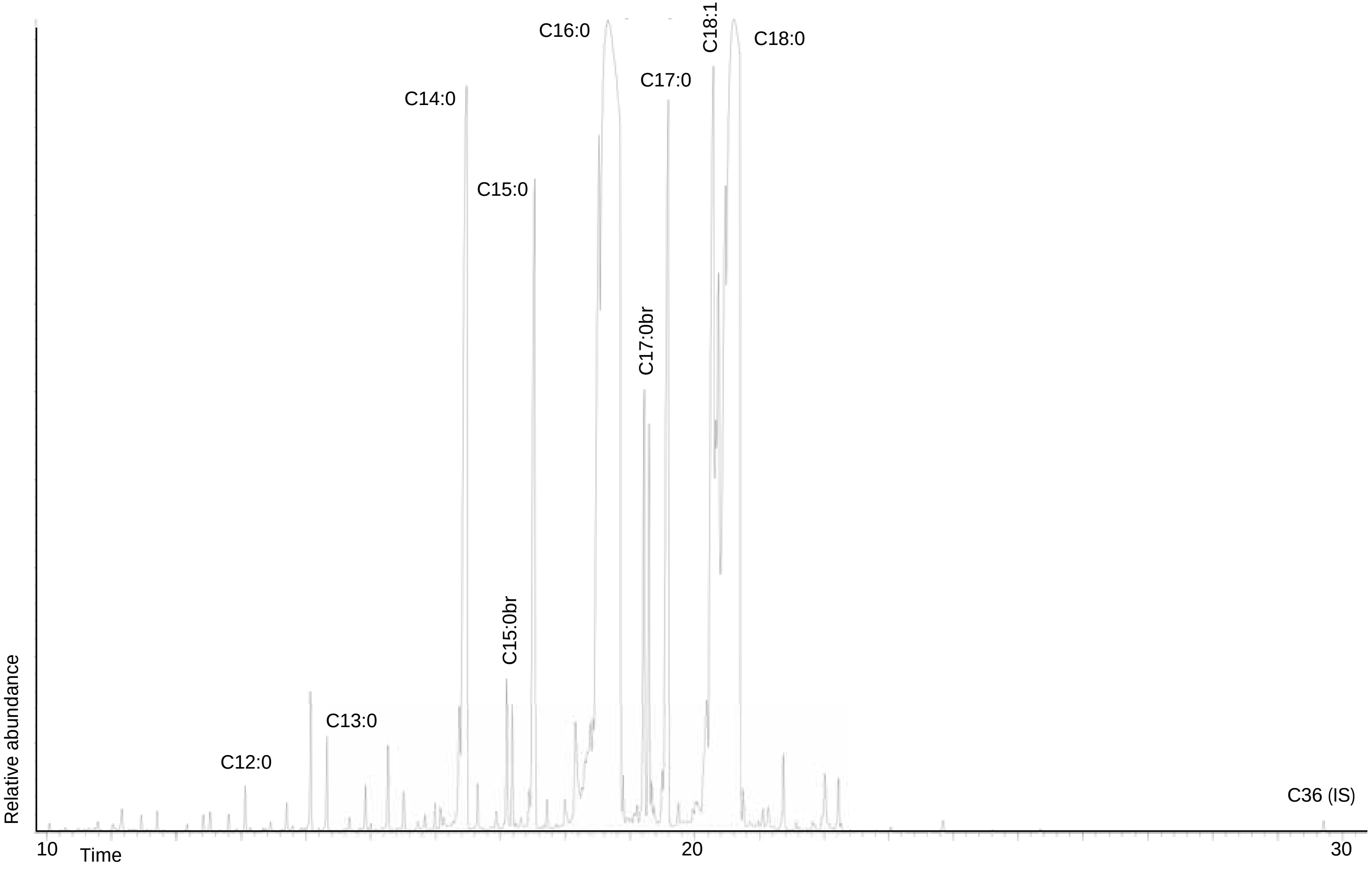
RA 9630


C16:0
C18:0
C18:1
C17:0
C14:0
C15:0
C17:0br
C20:0
C19:0
C15:0br
C12:0
C13:0
C22:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

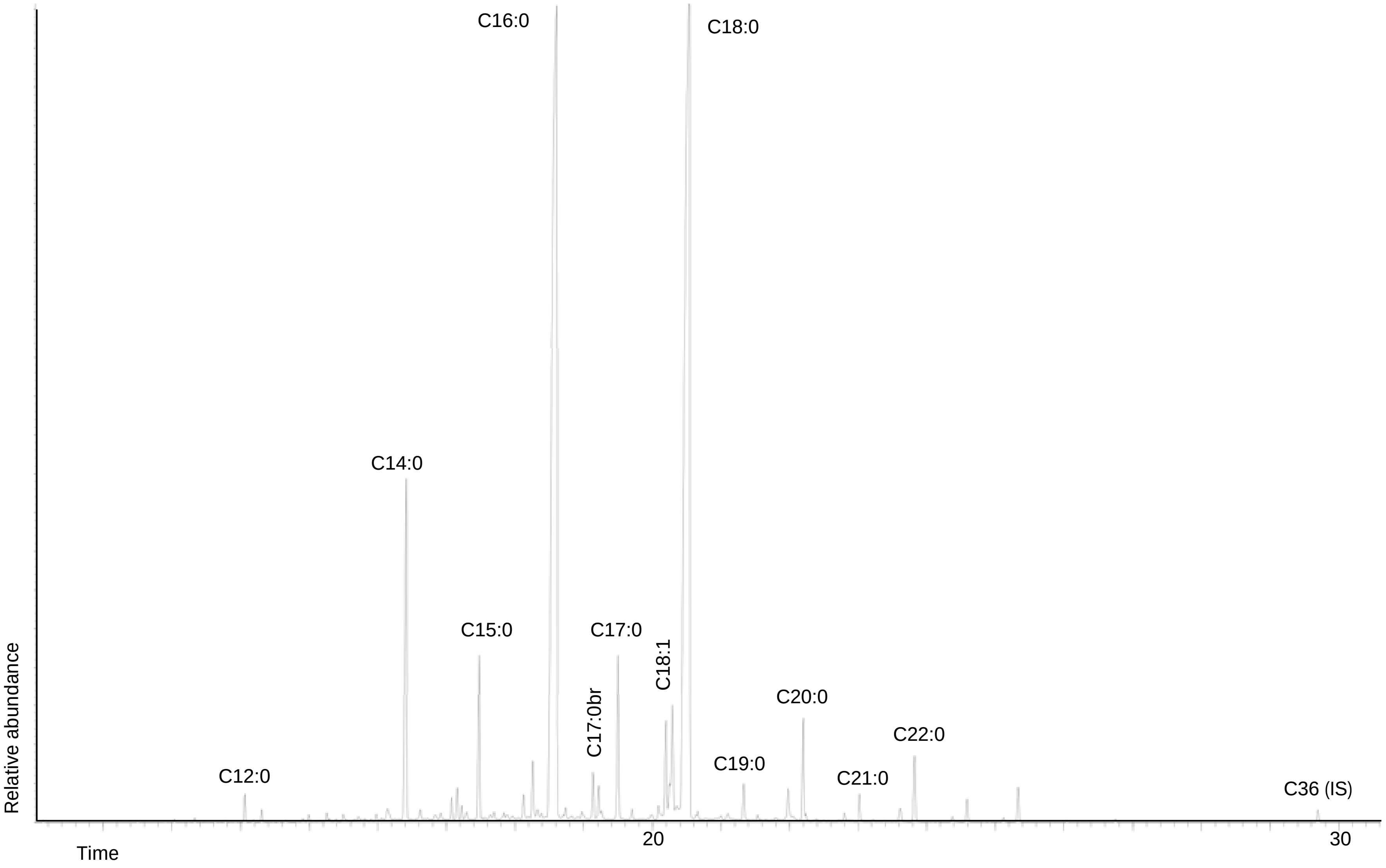
RA 9674


C16:0
C18:0
C17:0
C14:0
C15:0
C18:1
C20:0
C19:0
C12:0
C13:0
C22:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

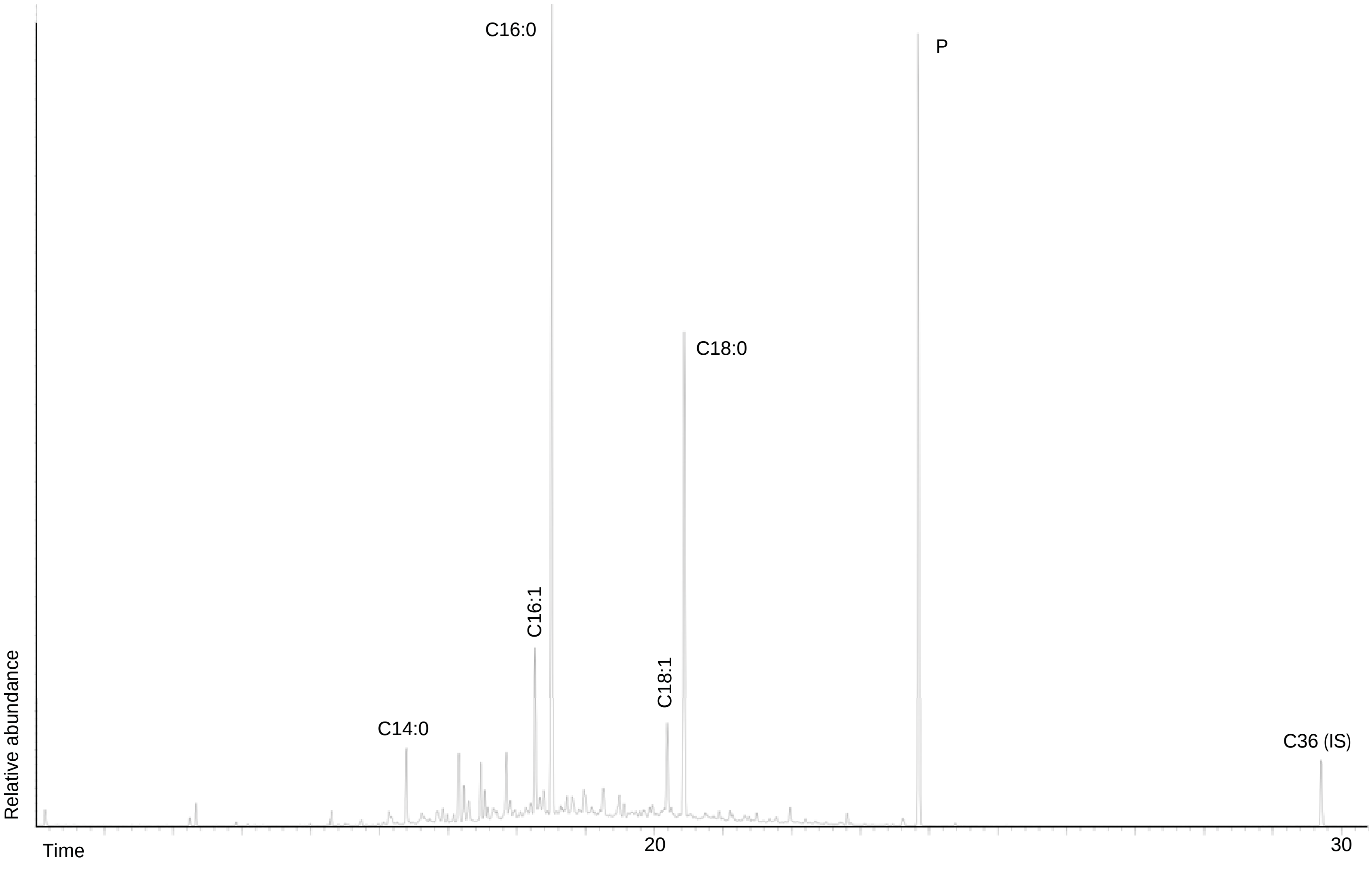
RA 9676


C16:0
C18:1
C18:0
C17:0
C14:0
C15:0
C17:0br
C15:0br
C13:0
C12:0
C36 (IS)
Relative abundance
10
Time
20
30

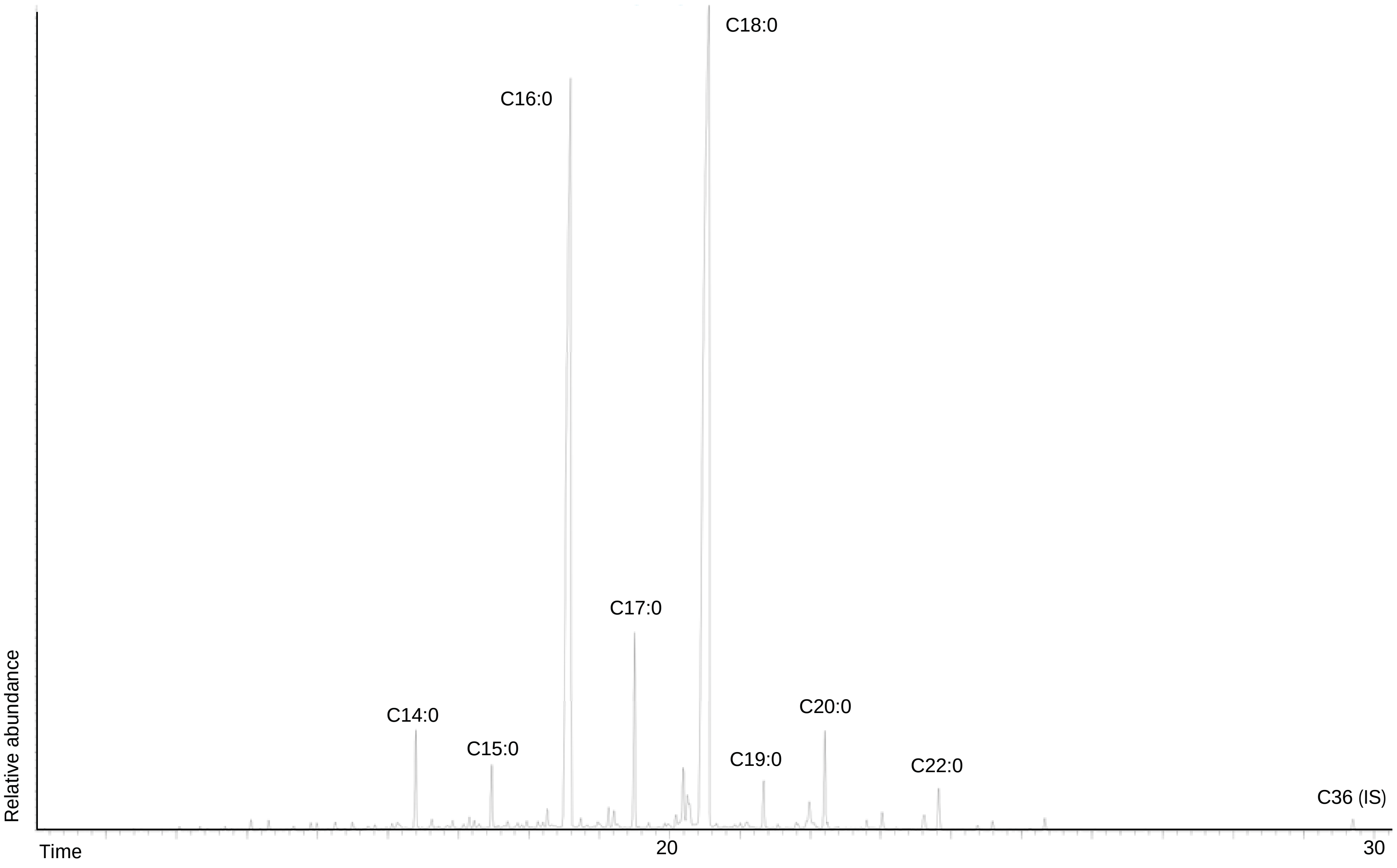
RA 9680


C16:0
C18:0
C14:0
C15:0
C17:0
C18:1
C17:0br
C20:0
C22:0
C12:0
C19:0
C21:0
C36 (IS)
Relative abundance
Time
20
30

RA 9684

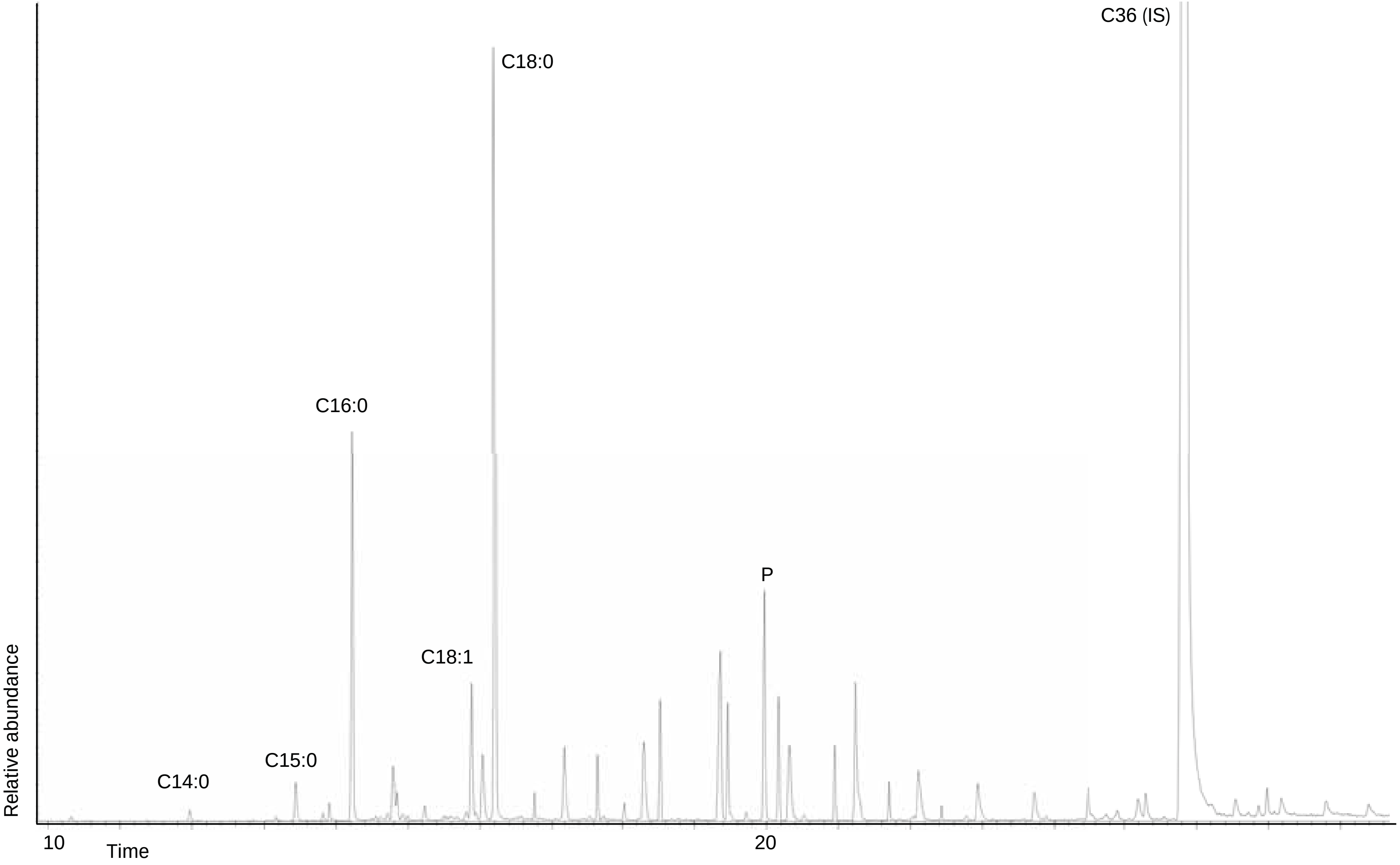

C16:0
P
C18:0
C16:1
C18:1
C14:0
C36 (IS)
Relative abundance
Time
20
30

RA 9734

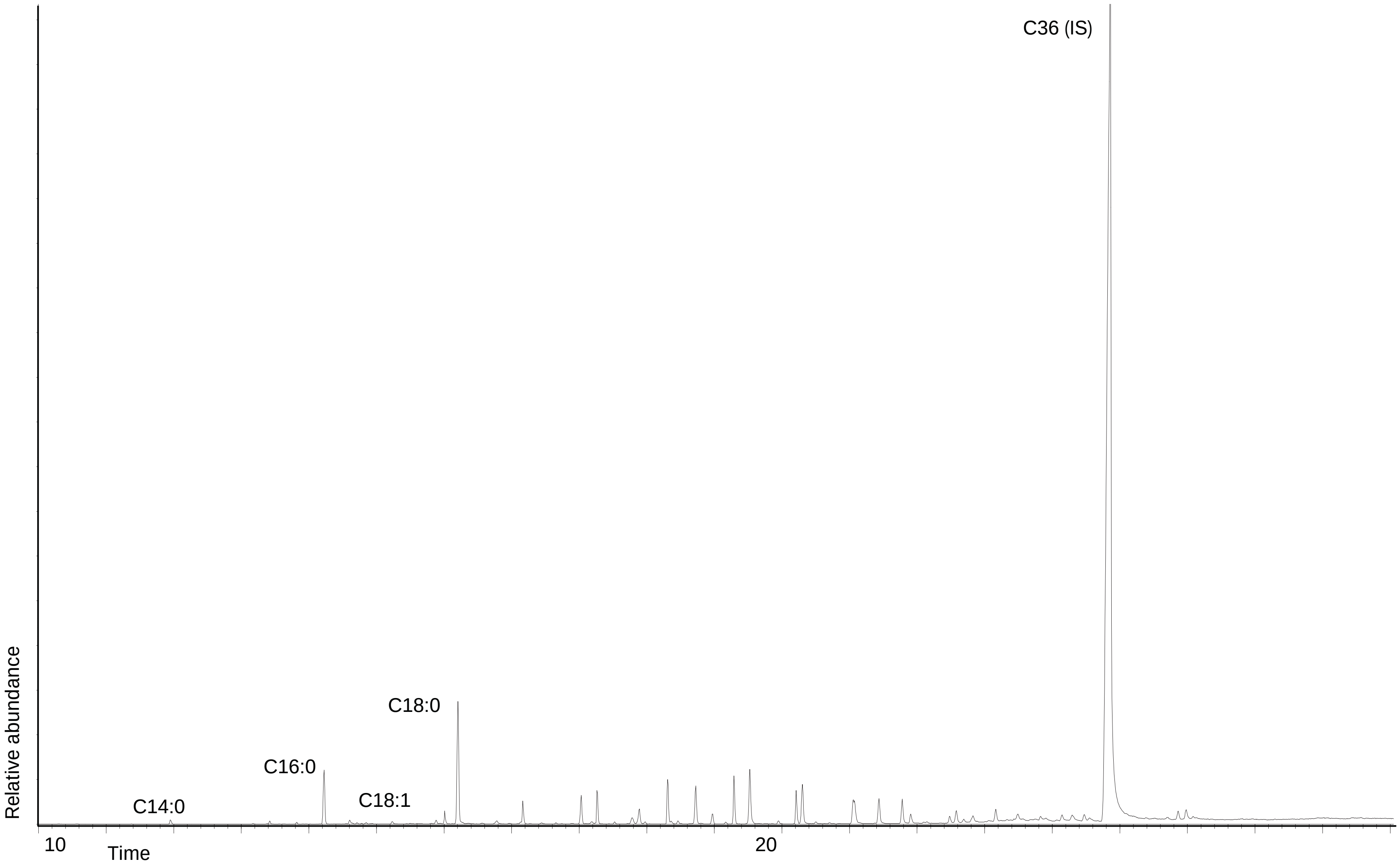

C18:0
C16:0
C17:0
C14:0
C20:0
C15:0
C19:0
C22:0
C36 (IS)
Relative abundance
Time
20
30

Cova de Sant Llorenç

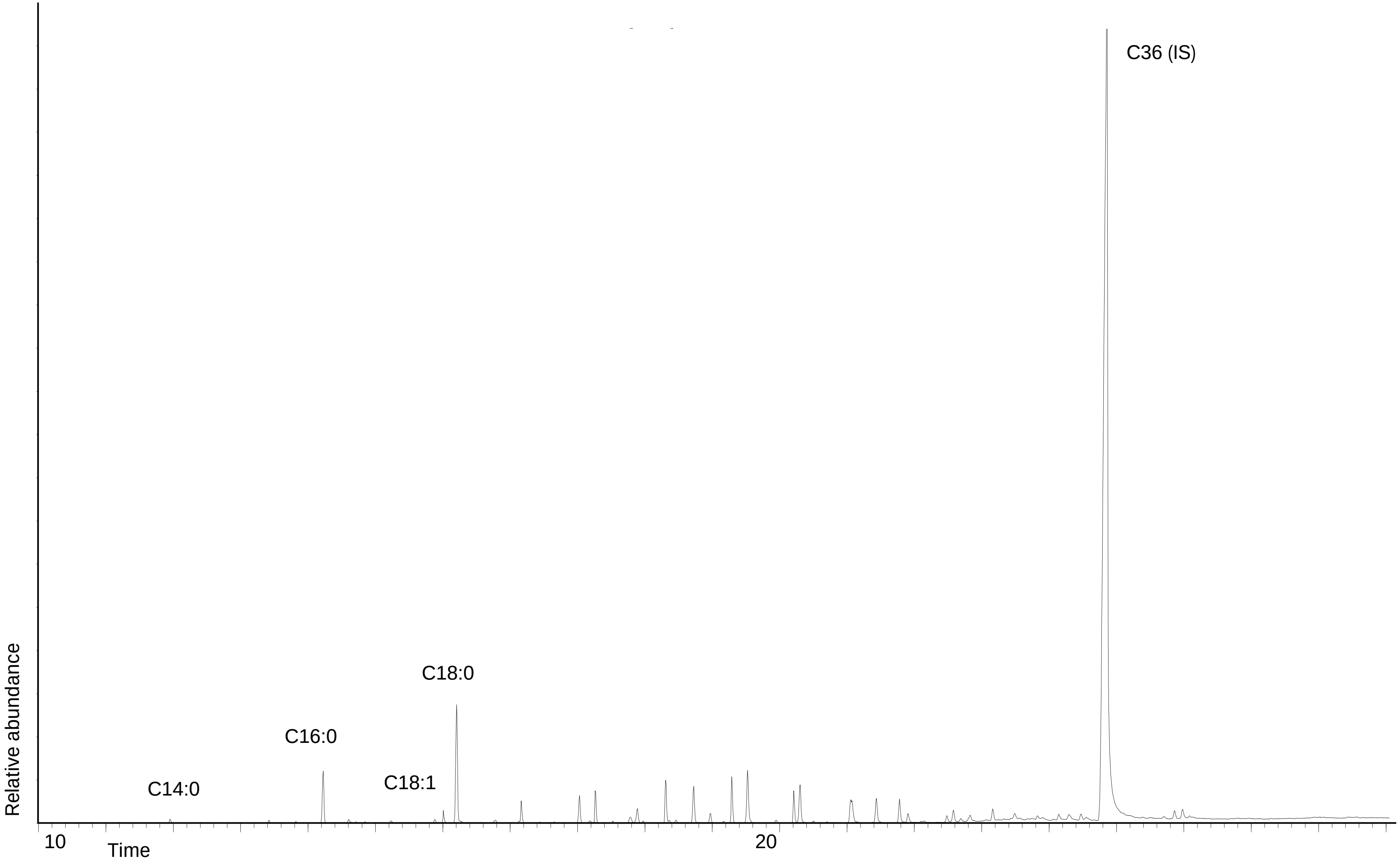
CSLL13


C36 (IS)
C18:0
C16:0
P
C18:1
C15:0
C14:0
Relative abundance
10
20
Time

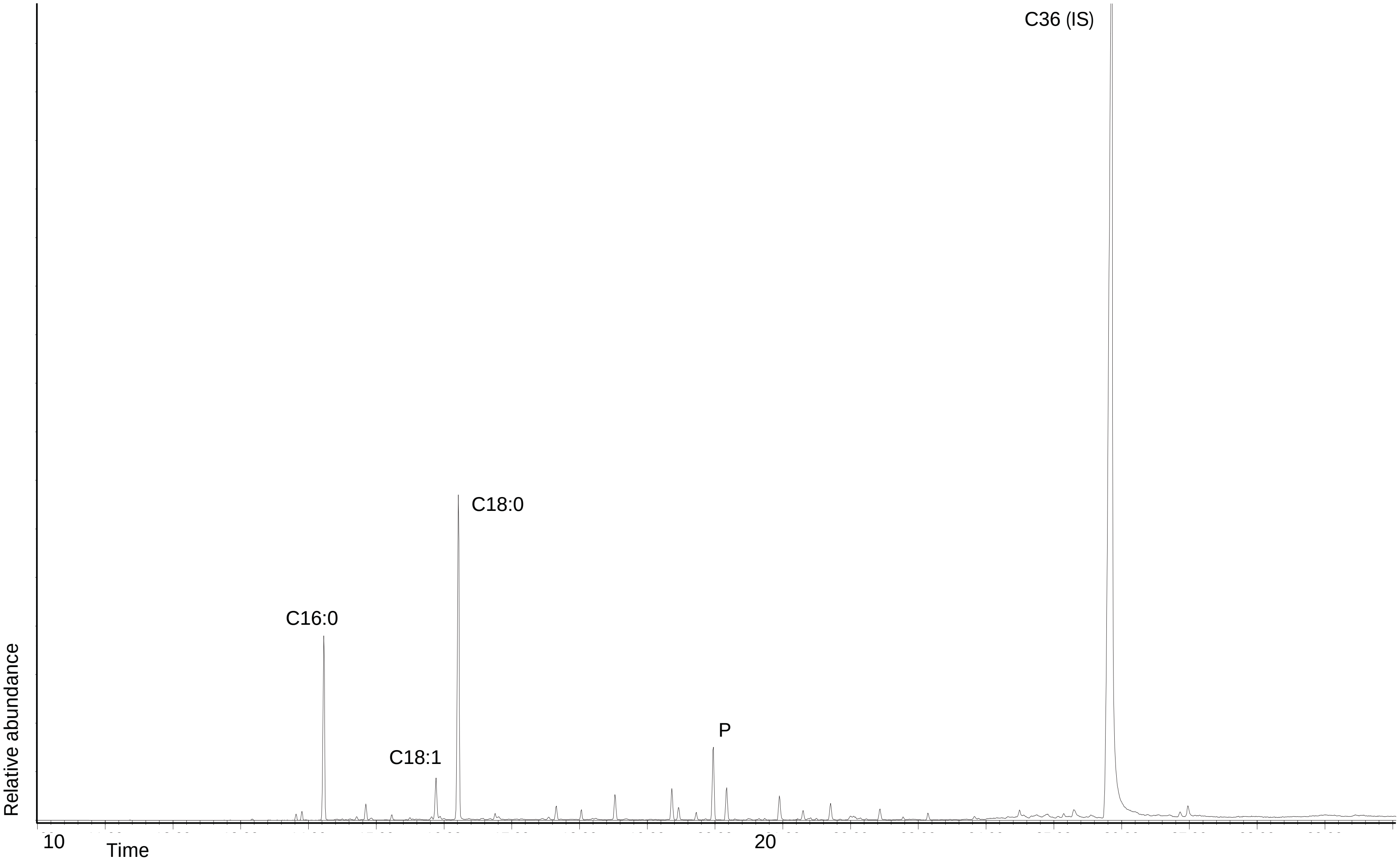
CSLL14



CSLL15

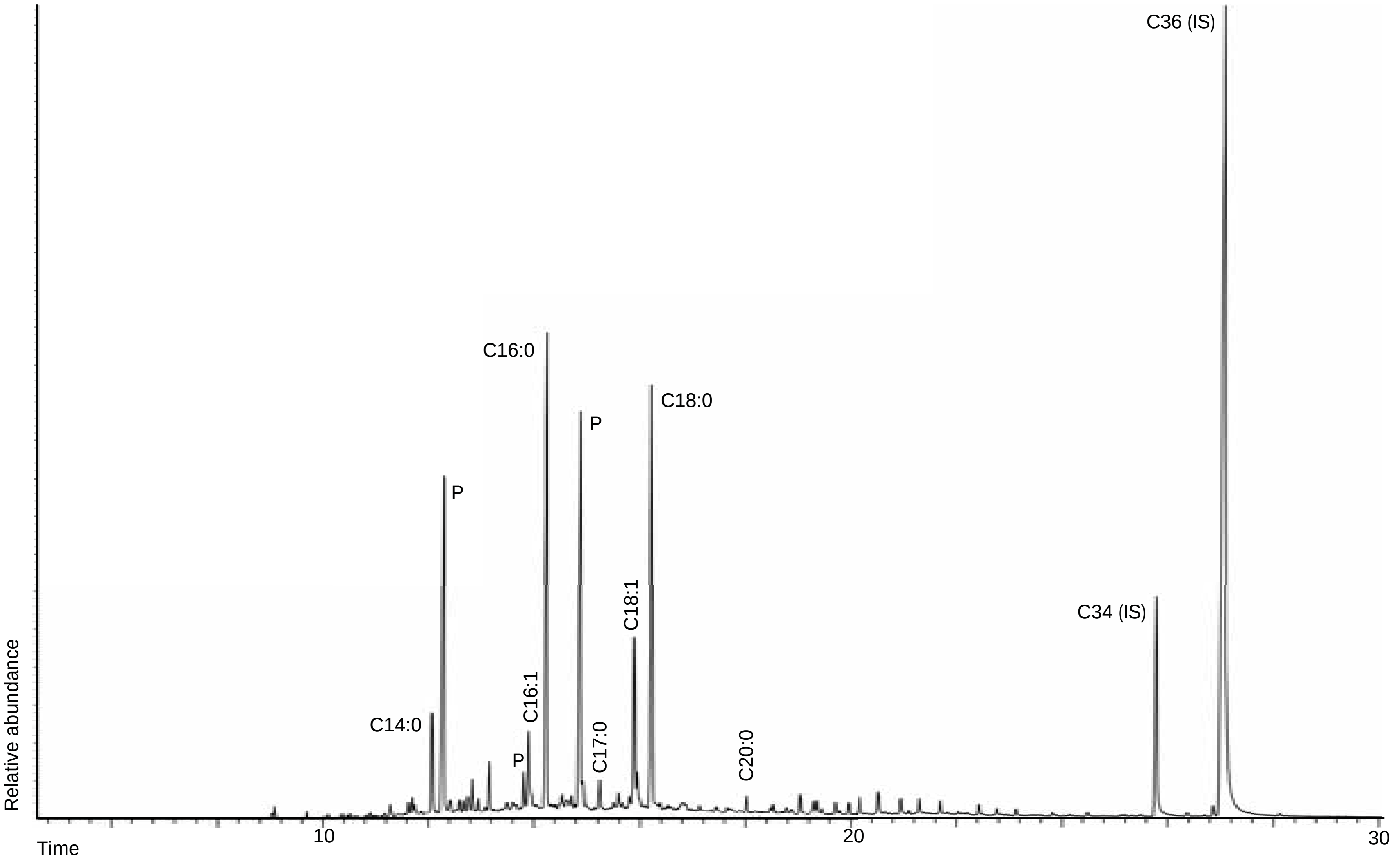


CSLL16



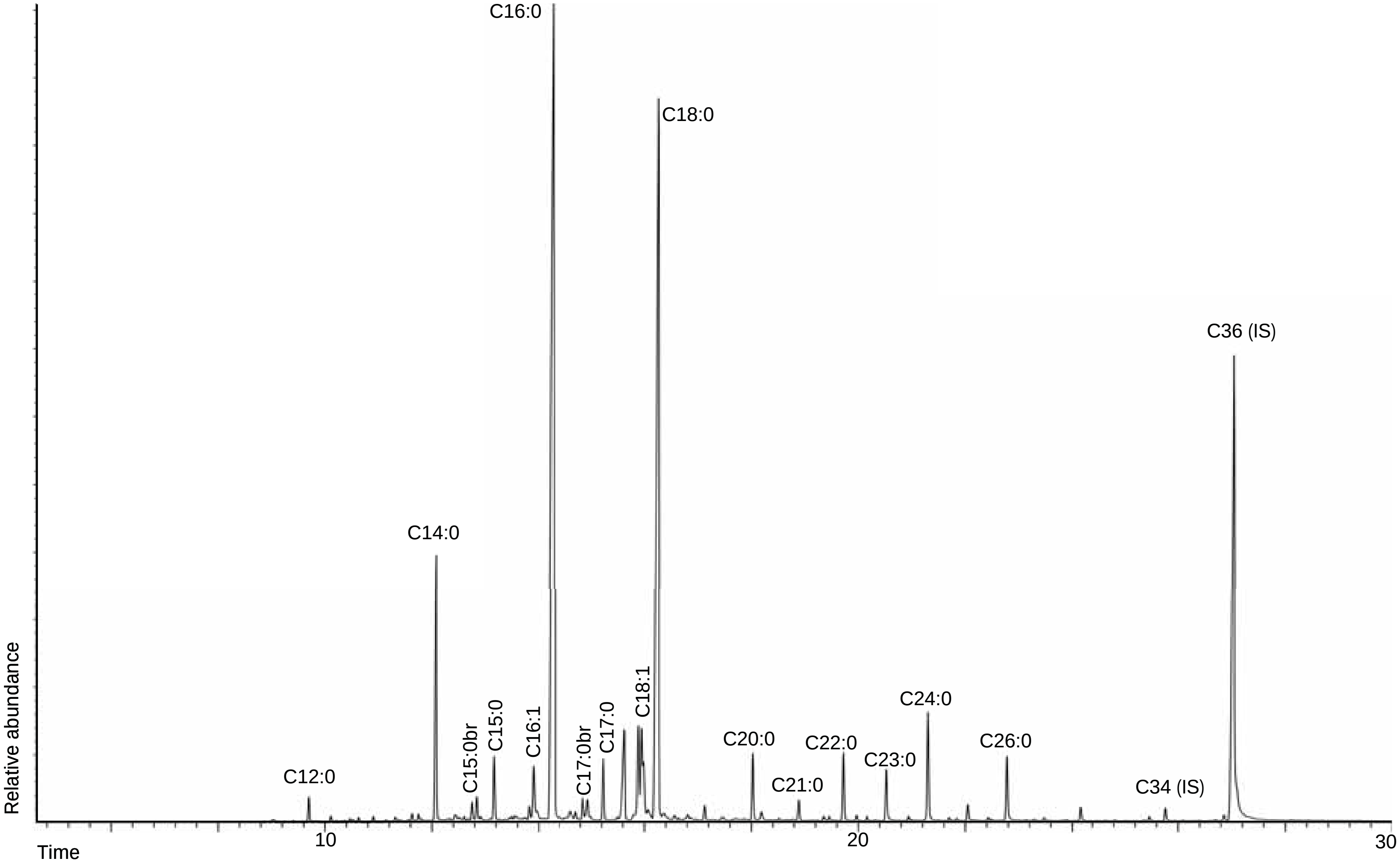
Can Sadurní

CS1

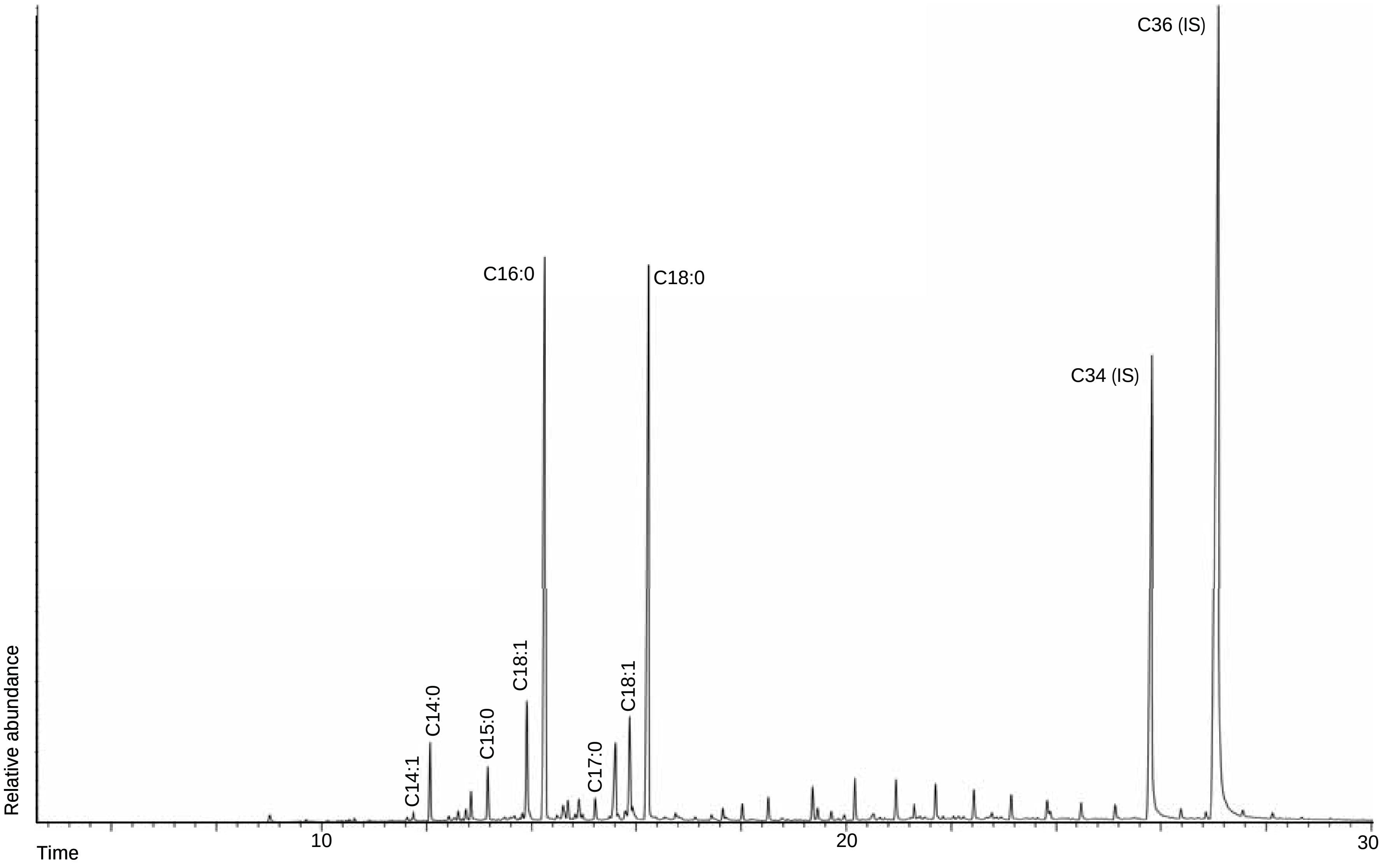


C36 (IS)
C16:0
C18:0
P
P
C34 (IS)
C18:1
C16:1
C14:0
P
C17:0
C20:0
Relative abundance
Time
10
20
30

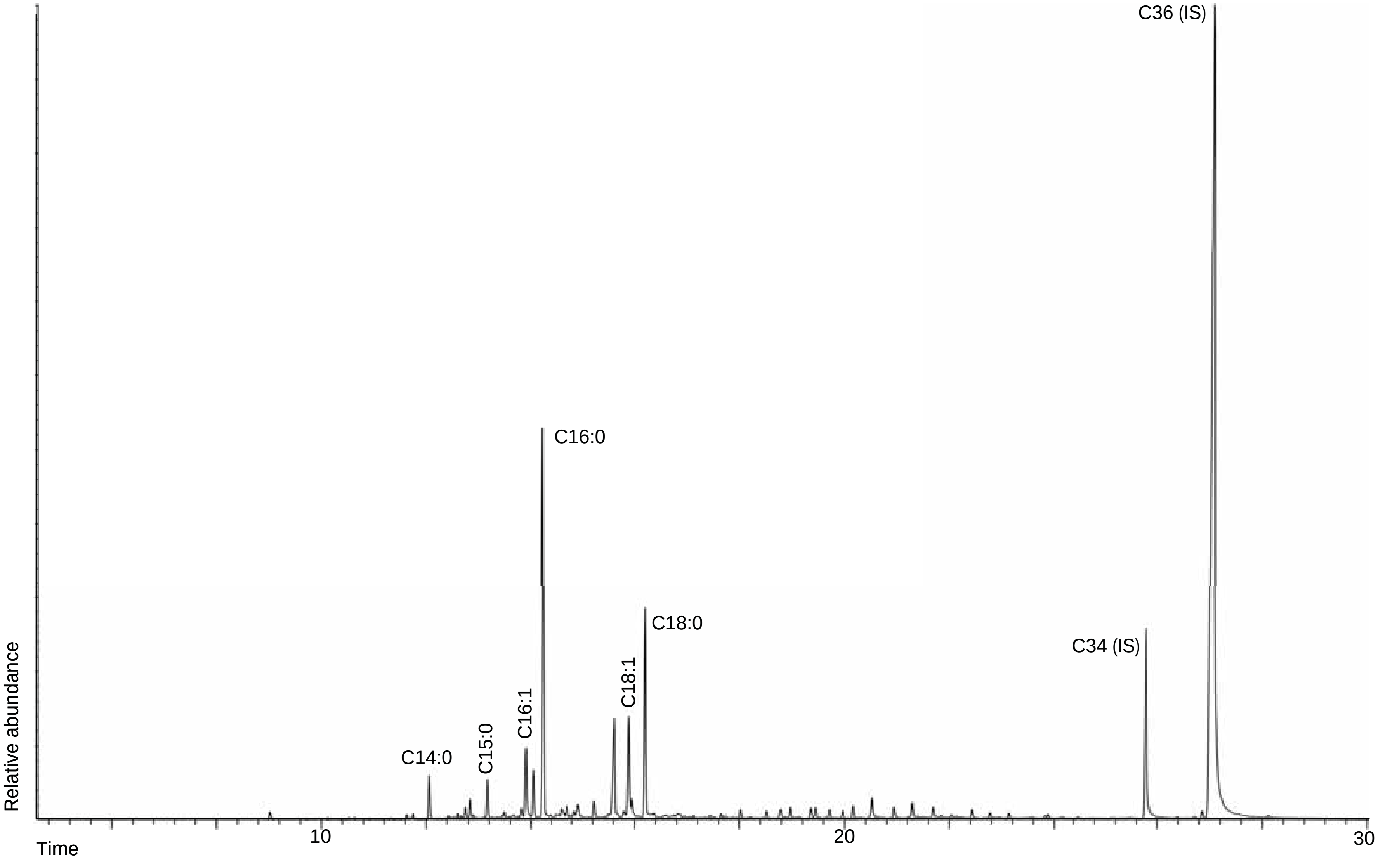
CS2


C16:0
C18:0
C14:0
C36 (IS)
C12:0
C15:0br
C15:0
C16:1
C17:0br
C17:0
C18:1
C20:0
C21:0
C22:0
C23:0
C24:0
C26:0
C34 (IS)
Relative abundance
Time
10
20
30

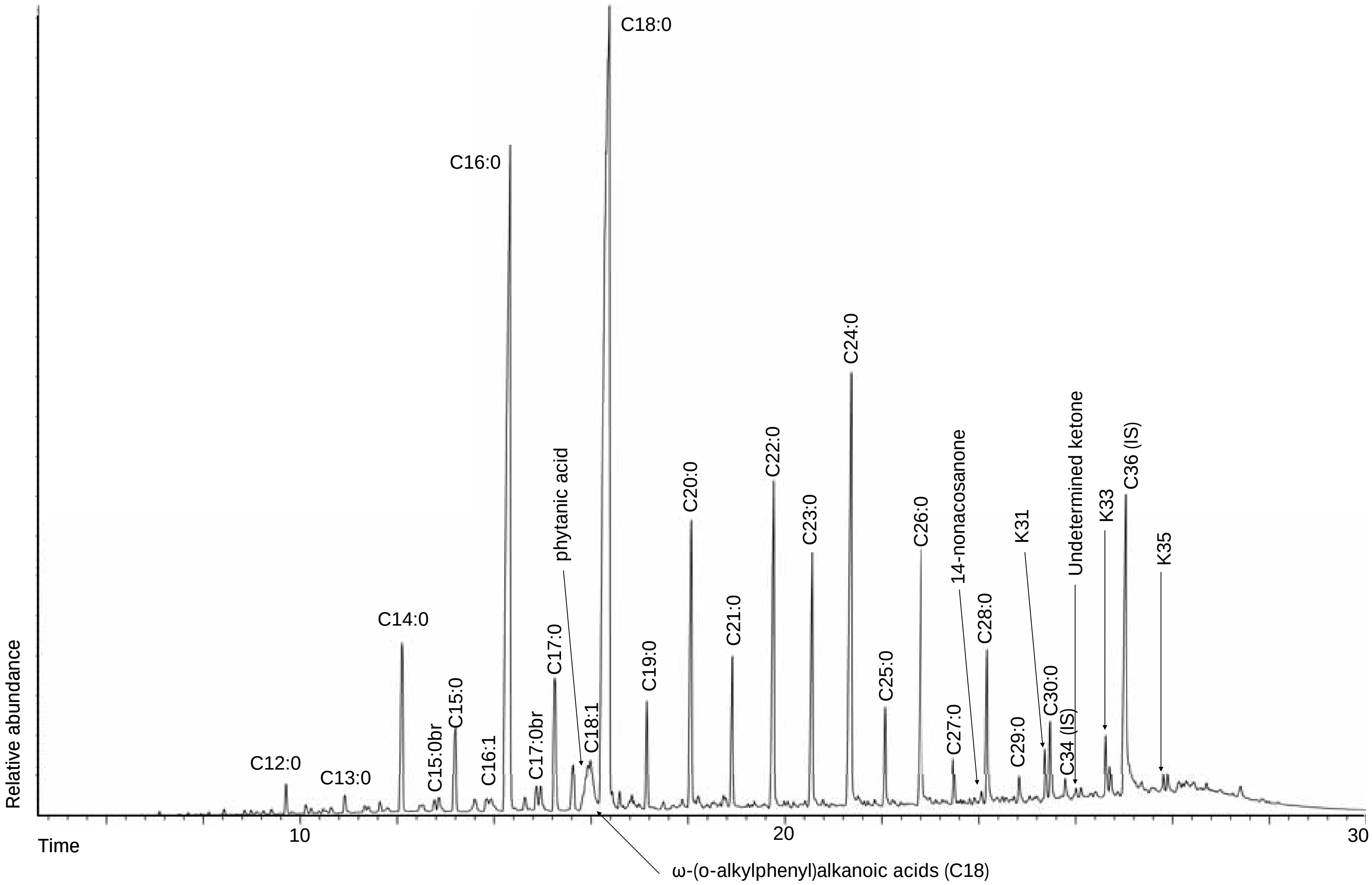
CS3


C36 (IS)
C16:0
C18:0
C34 (IS)
C18:1
C18:1
C14:0
C15:0
C17:0
C14:1
Relative abundance
Time
10
20
30

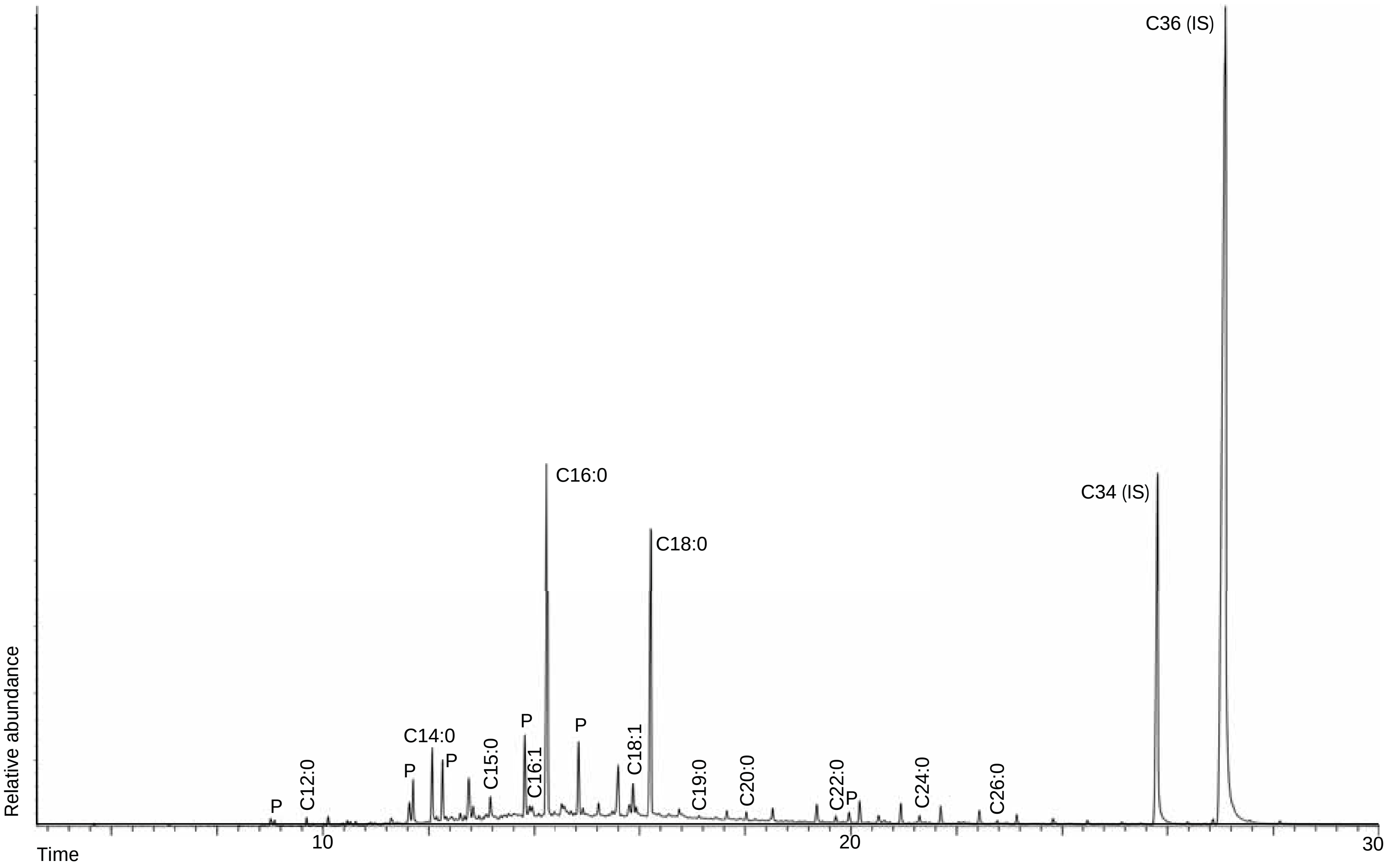
CS4


C36 (IS)
C16:0
C18:0
C34 (IS)
C18:1
C16:1
C15:0
C14:0
Relative abundance
Time
10
20
30

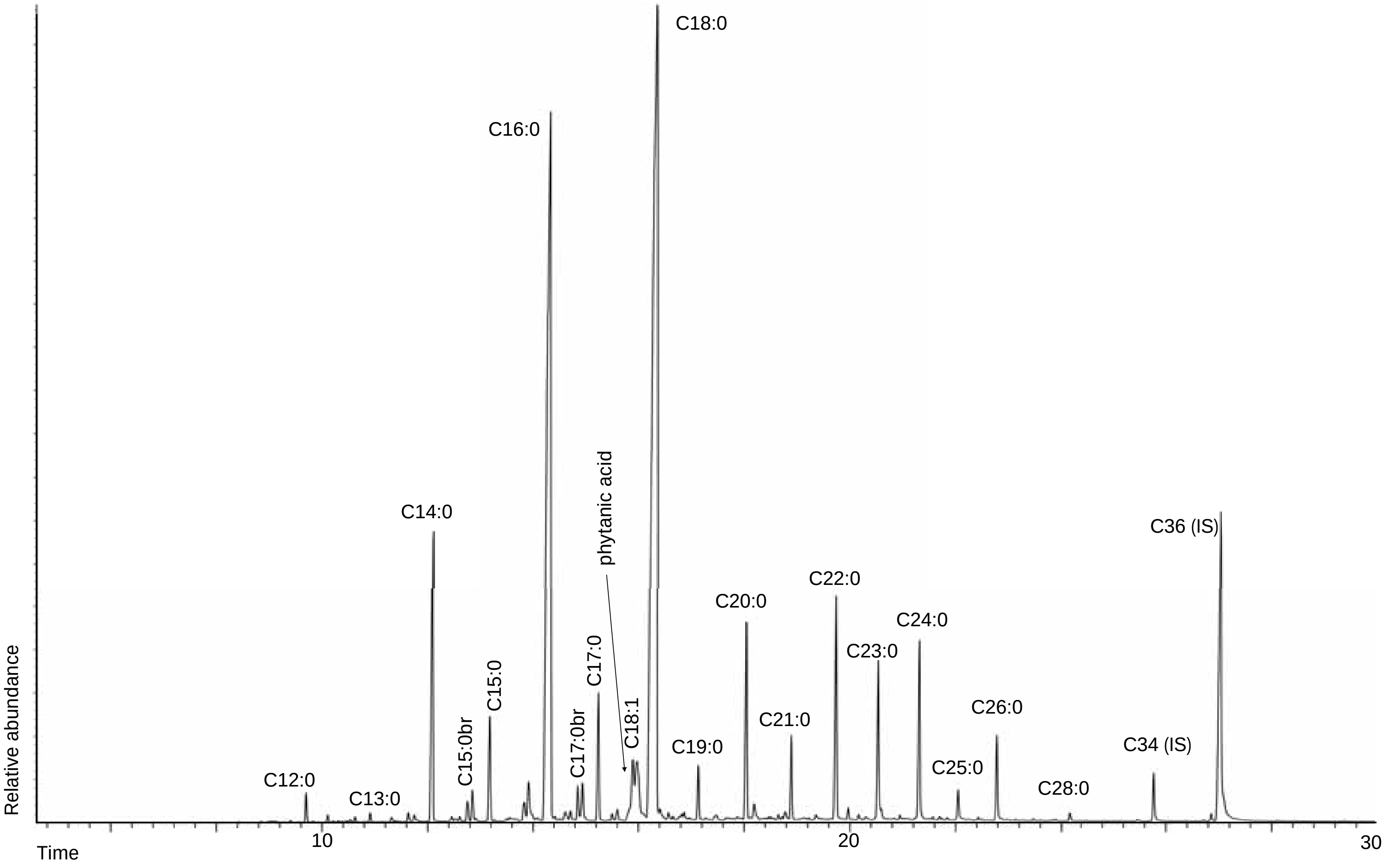
CS5


Relative abundance
Time
10
20
30
C12:0
C13:0
C14:0
C15:0br
C15:0
C16:1
C16:0
C17:0br
C17:0
phytanic acid
C18:1
C18:0
ω-(o-alkylphenyl)alkanoic acids (C18)
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
C27:0
14-nonacosanone
C28:0
C29:0
K31
C30:0
C34 (IS)
Undetermined ketone
K33
C36 (IS)
K35

CS6

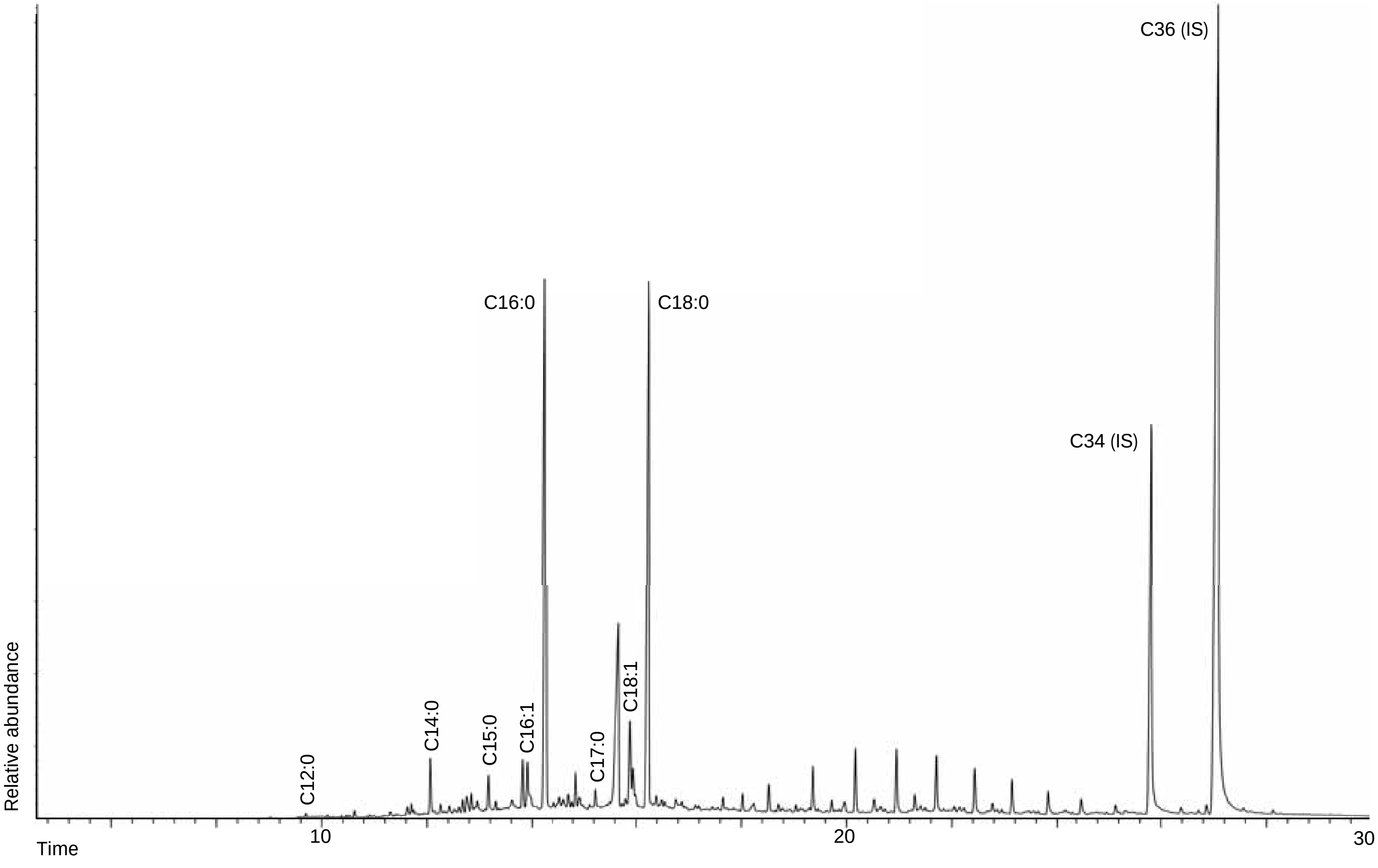

C36 (IS)
C34 (IS)
C16:0
C18:0
P
C12:0
P
C14:0
P
C15:0
P
C16:1
P
C18:1
C19:0
C20:0
C22:0
P
C24:0
C26:0
Relative abundance
Time
10
20
30

CS7

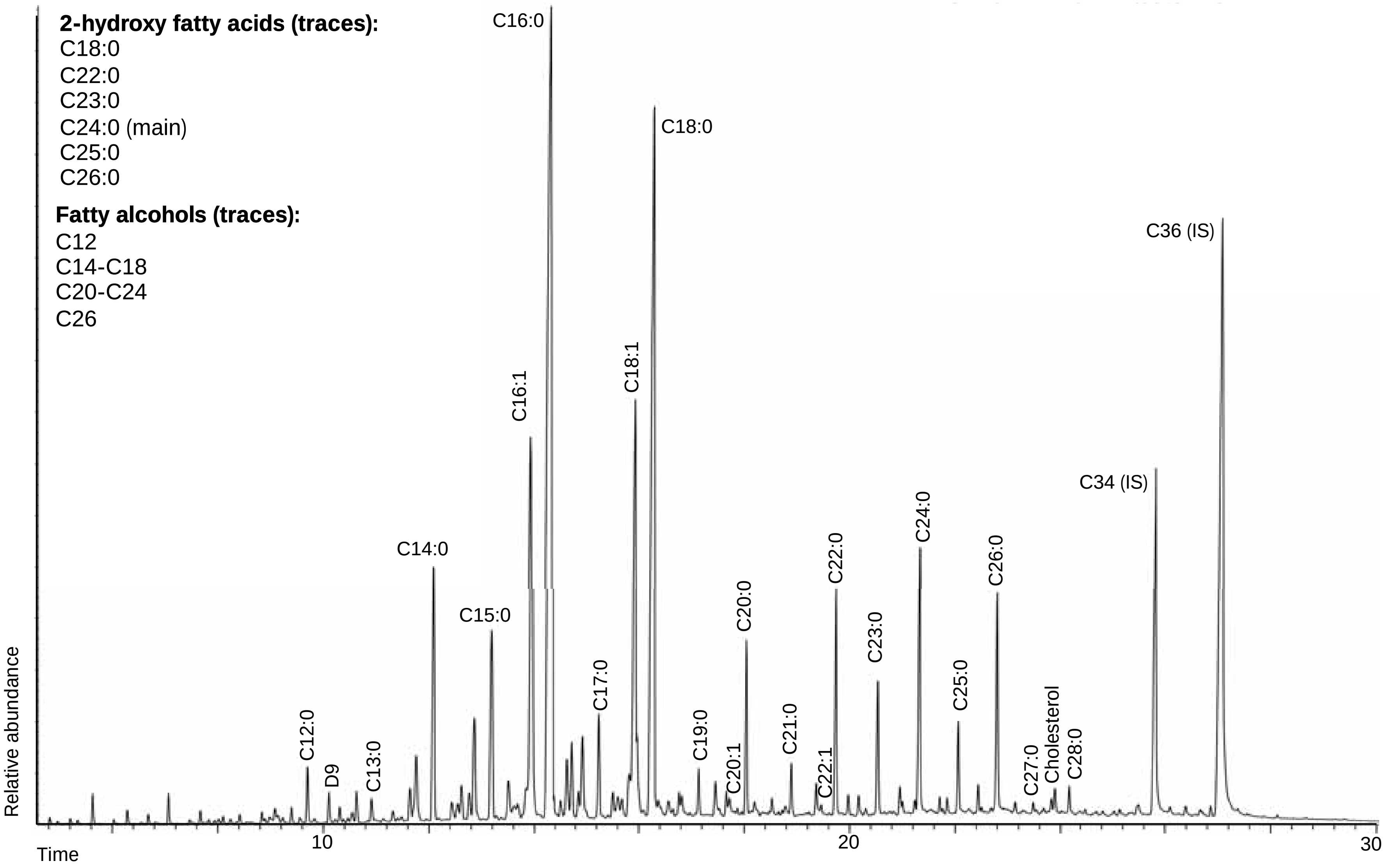


C18:0
C16:0
phytanic acid
C14:0
C36 (IS)
C22:0
C20:0
C24:0
C23:0
C15:0
C17:0
C15:0br
C17:0br
C18:1
C21:0
C26:0
C19:0
C34 (IS)
C25:0
C12:0
C28:0
C13:0
Relative abundance
Time
10
20
30

CS8


C36 (IS)
C16:0
C18:0
C34 (IS)
C18:1
C14:0
C16:1
C15:0
C17:0
C12:0
Relative abundance
Time
10
20
30

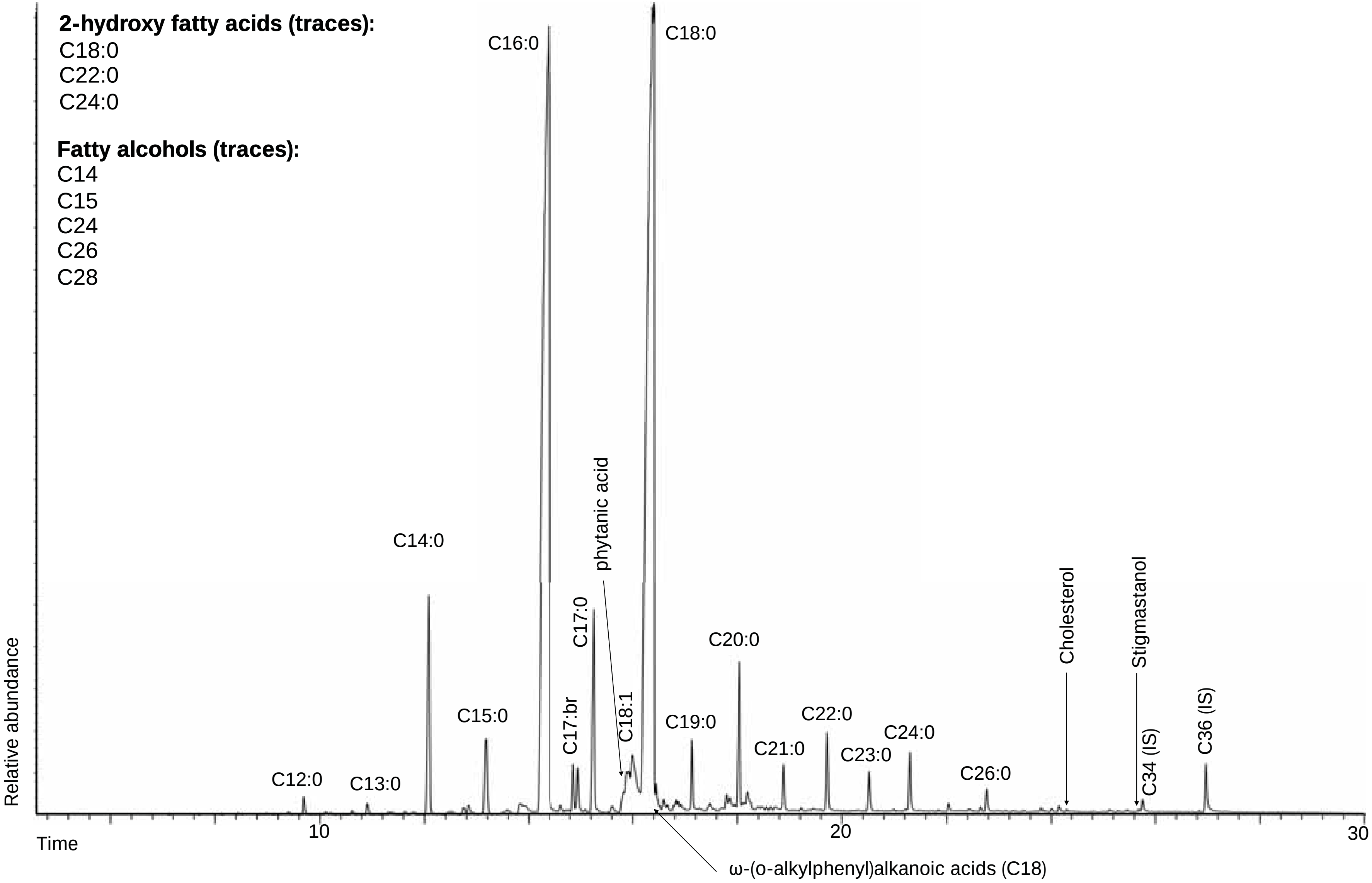
CS9



2-hydroxy fatty acids (traces):
C18:0
C22:0
C23:0
C24:0 (main)
C25:0
C26:0

Fatty alcohols (traces):
C12
C14-C18
C20-C24
C26

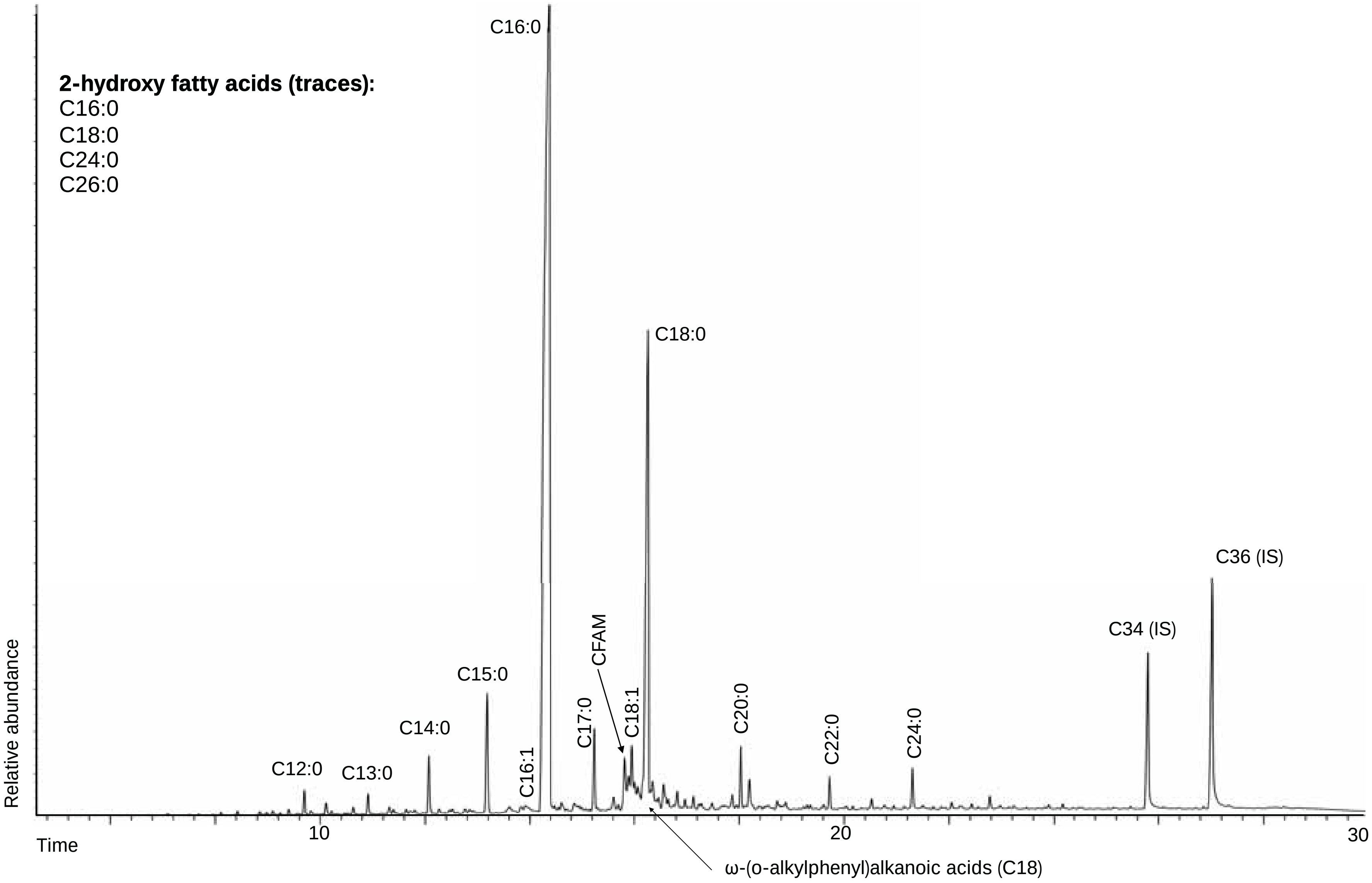
CS10



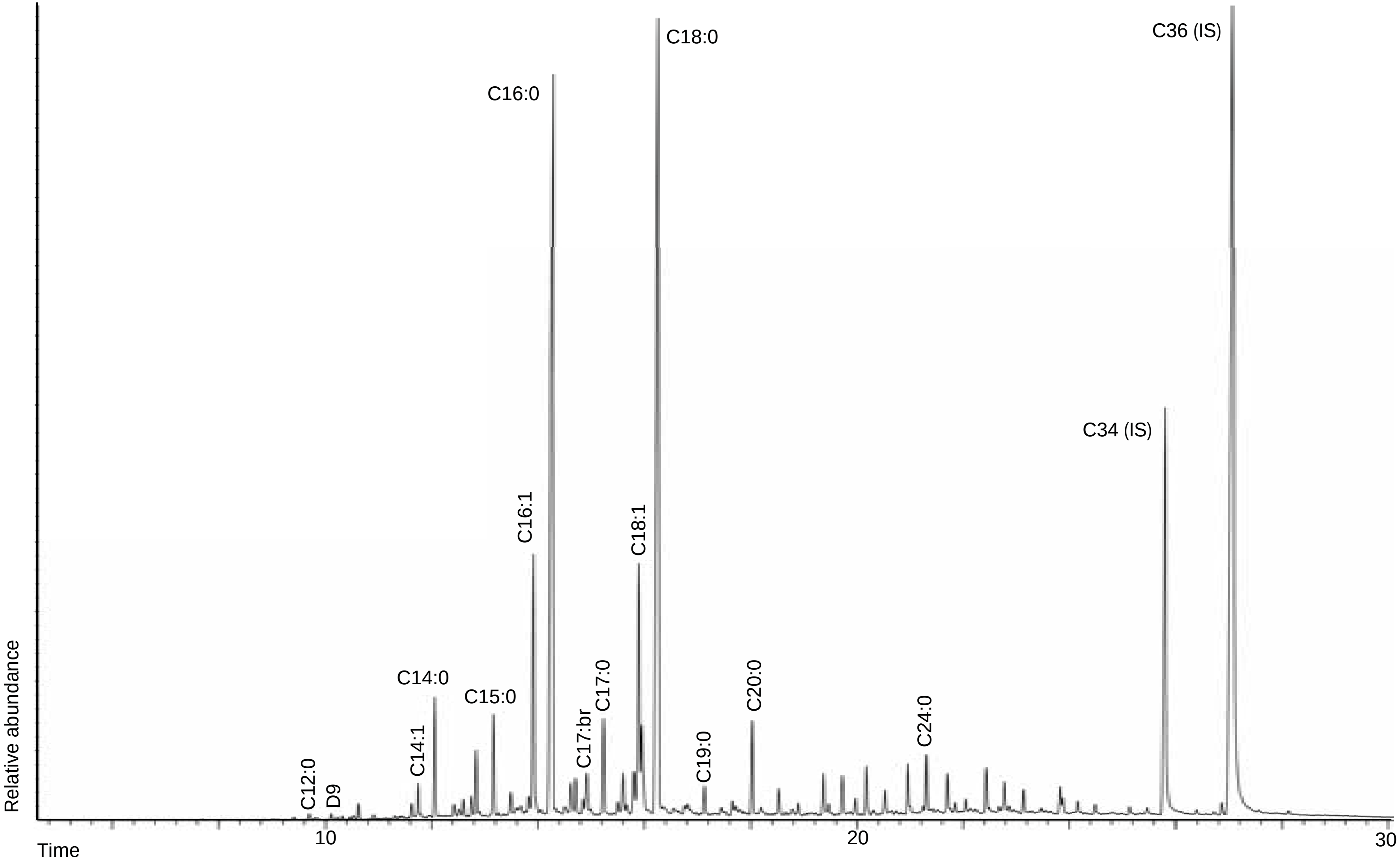
2-hydroxy fatty acids (traces):
C18:0
C22:0
C24:0

Fatty alcohols (traces):
C14
C15
C24
C26
C28

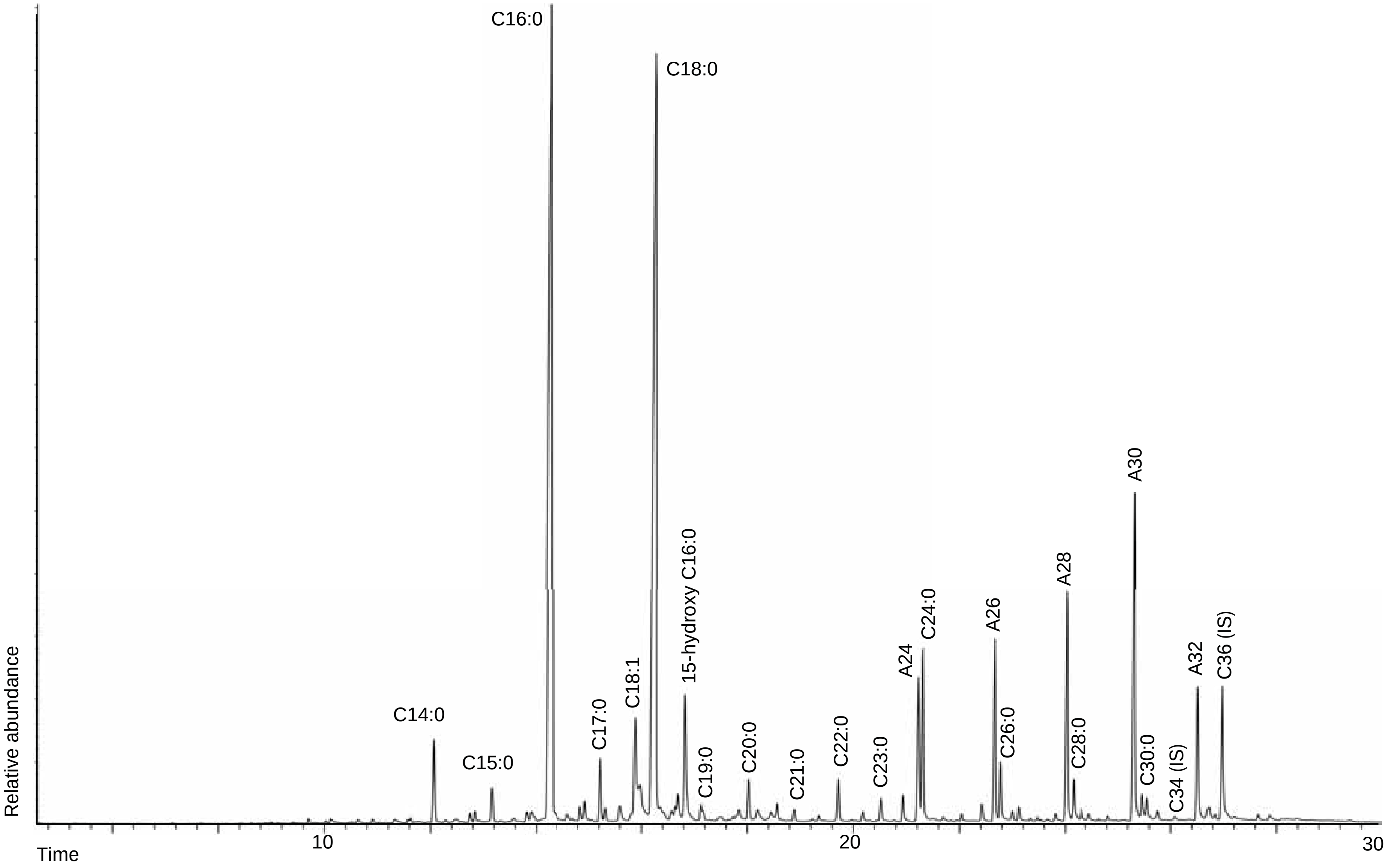
CS12


2-hydroxy fatty acids (traces):
C16:0
C18:0
C24:0
C26:0
C16:0
C18:0
C36 (IS)
C34 (IS)
CFAM
C15:0
C14:0
C17:0
C18:1
C20:0
C22:0
C24:0
C16:1
C12:0
C13:0
Relative abundance
Time
10
20
30
ω-(o-alkylphenyl)alkanoic acids (C18)

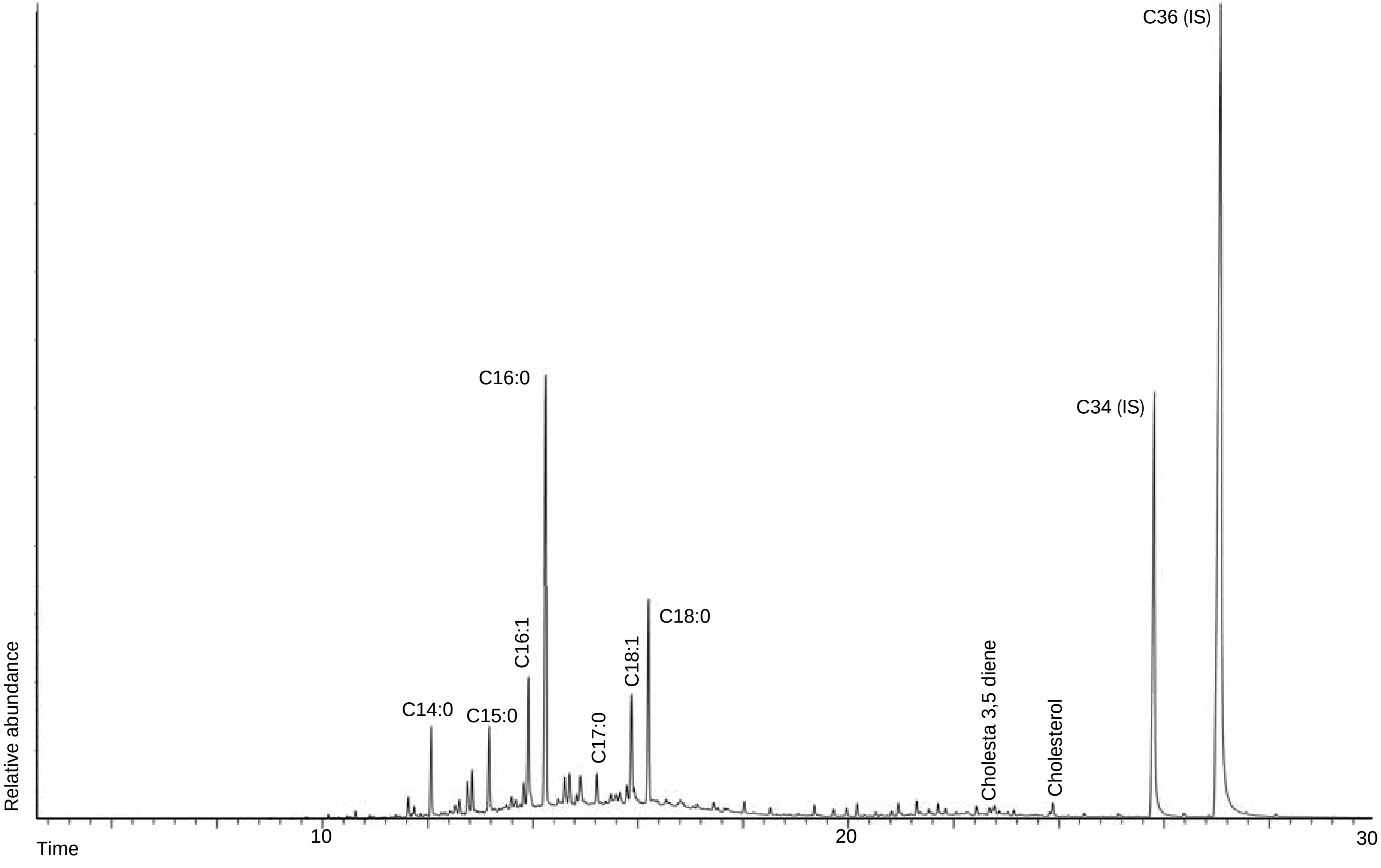
CS13


Relative abundance
Time
10
20
30
C12:0
D9
C14:1
C14:0
C15:0
C16:1
C16:0
C17:br
C17:0
C18:1
C18:0
C19:0
C20:0
C24:0
C34 (IS)
C36 (IS)

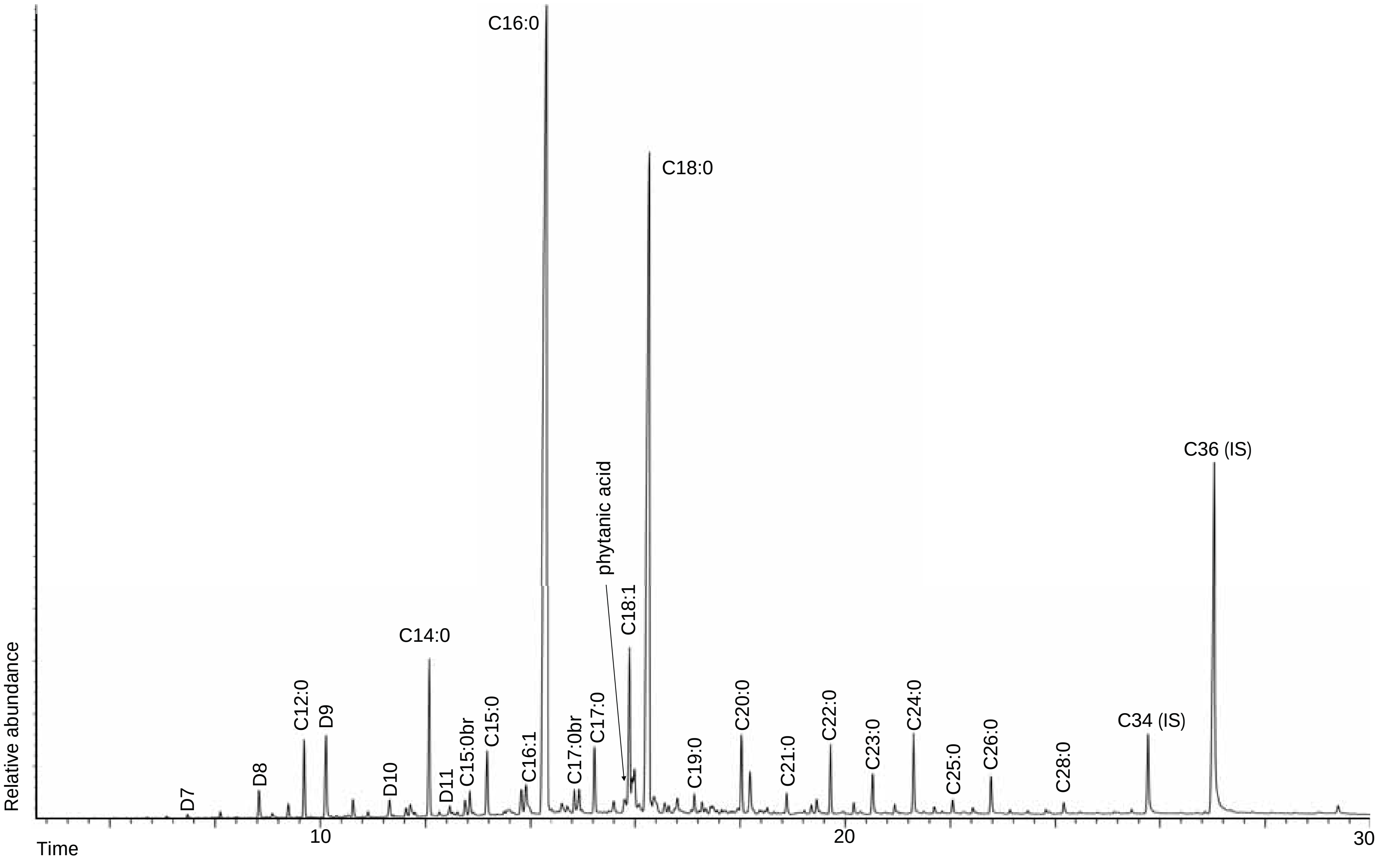
CS14


C16:0
C18:0
A30
15-hydroxy C16:0
A28
C24:0
A26
A24
C36 (IS)
A32
C18:1
C14:0
C17:0
C26:0
C28:0
C15:0
C20:0
C22:0
C30:0
C19:0
C23:0
C34 (IS)
C21:0
Relative abundance
Time
10
20
30

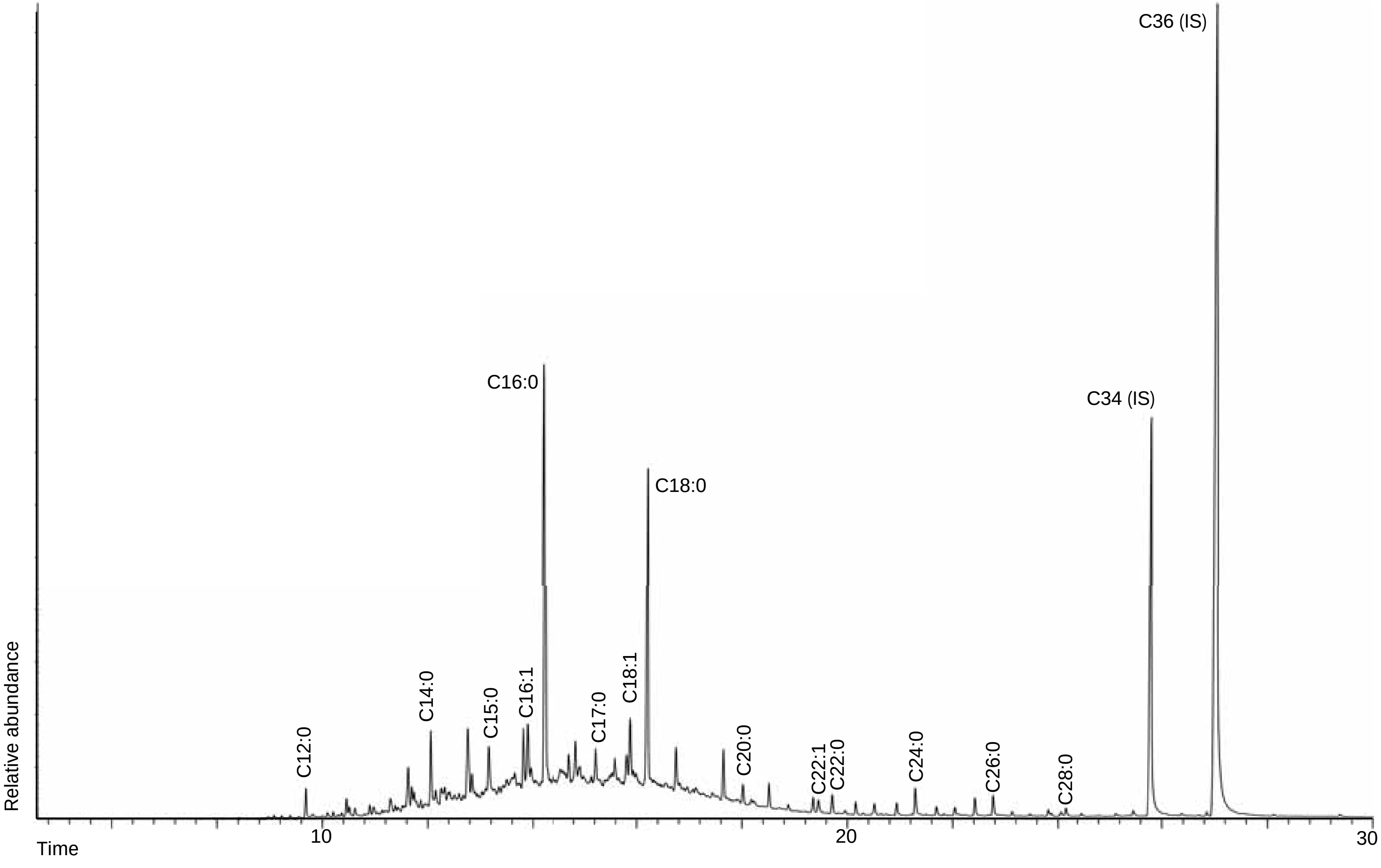
CS15


C36 (IS)
C34 (IS)
C16:0
C18:0
C16:1
C18:1
C14:0
C15:0
C17:0
Cholesta 3,5 diene
Cholesterol
Relative abundance
Time
10
20
30

CS16

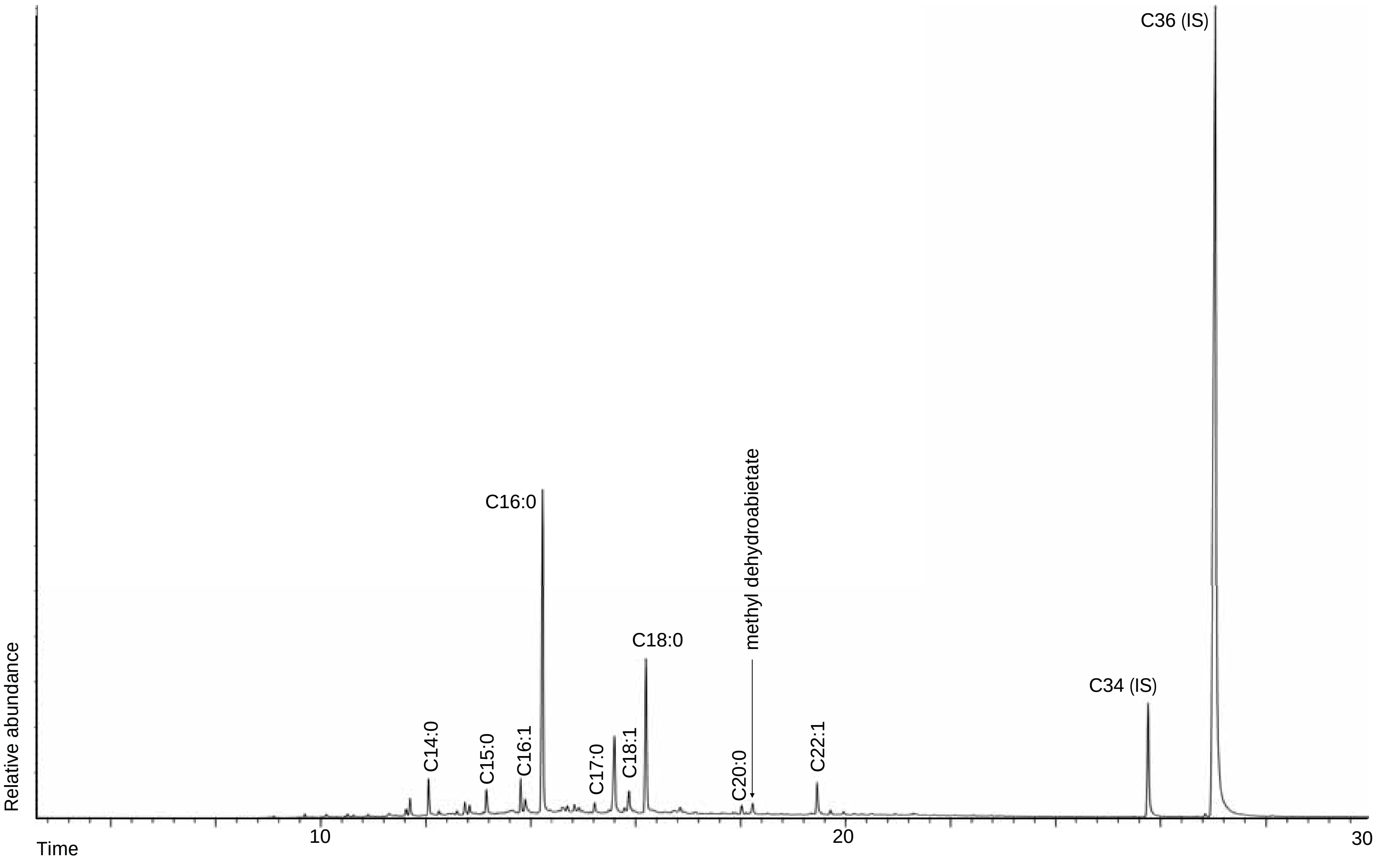

Relative abundance
Time
10
20
30
D7
D8
C12:0
D9
D10
C14:0
D11
C15:0br
C15:0
C16:1
C16:0
C17:0br
C17:0
phytanic acid
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
C28:0
C34 (IS)
C36 (IS)

CS17

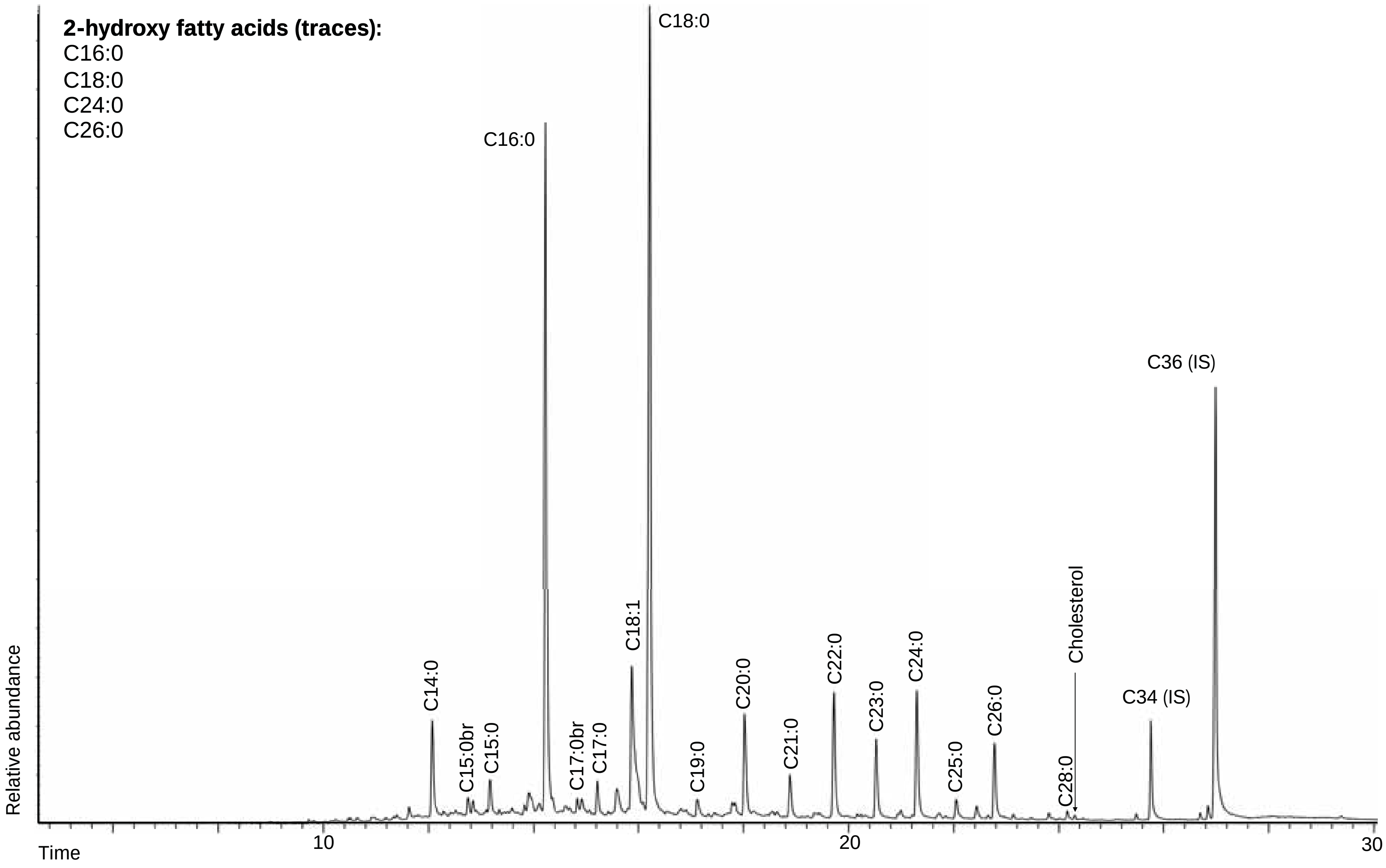


C36 (IS)
C34 (IS)
C16:0
C18:0
C12:0
C14:0
C15:0
C16:1
C17:0
C18:1
C20:0
C22:1
C22:0
C24:0
C26:0
C28:0
Relative abundance
Time
10
20
30

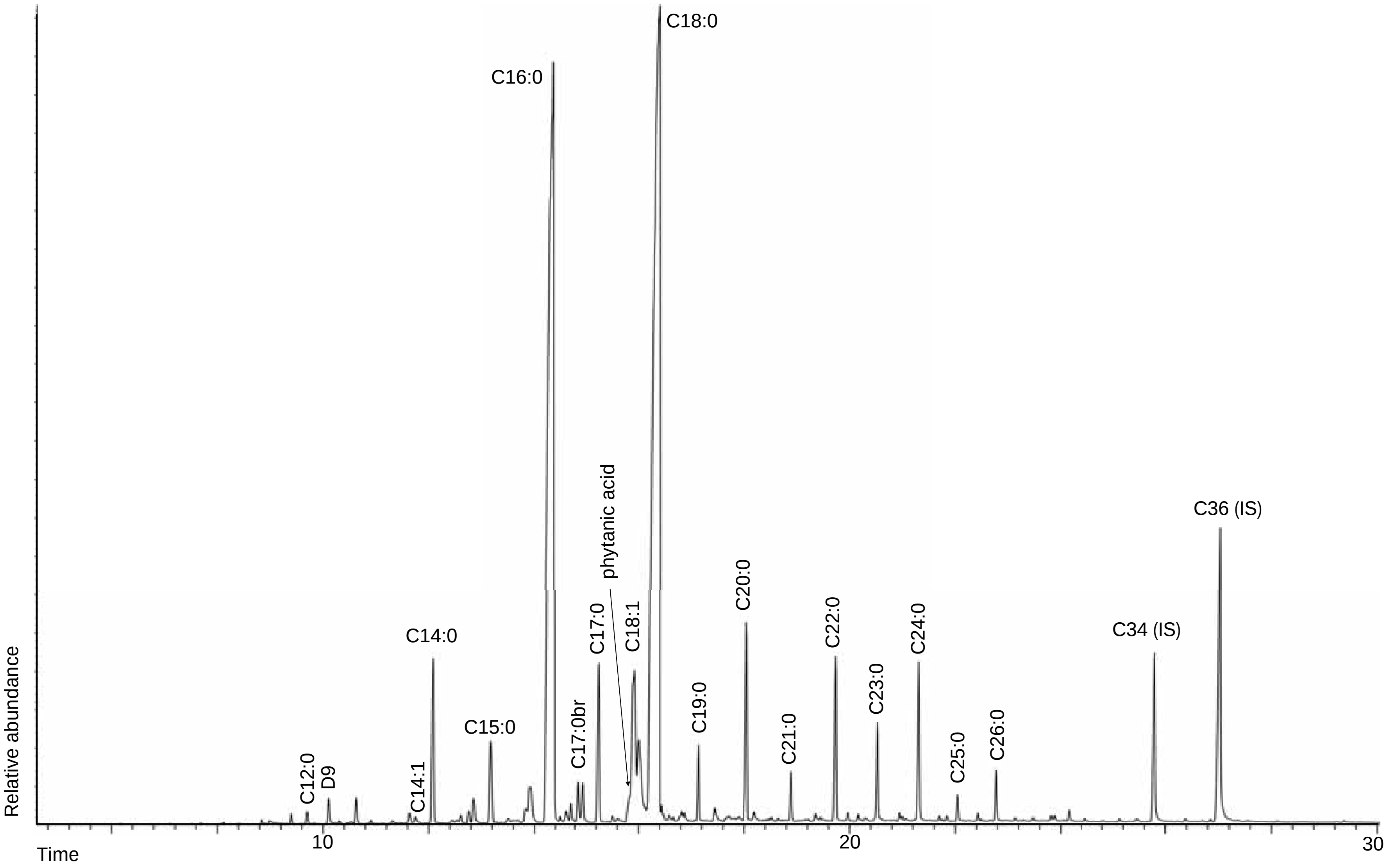
CS18


C36 (IS)
C16:0
C18:0
methyl dehydroabietate
C34 (IS)
C14:0
C15:0
C16:1
C17:0
C18:1
C20:0
C22:1
Relative abundance
Time
10
20
30

CS19

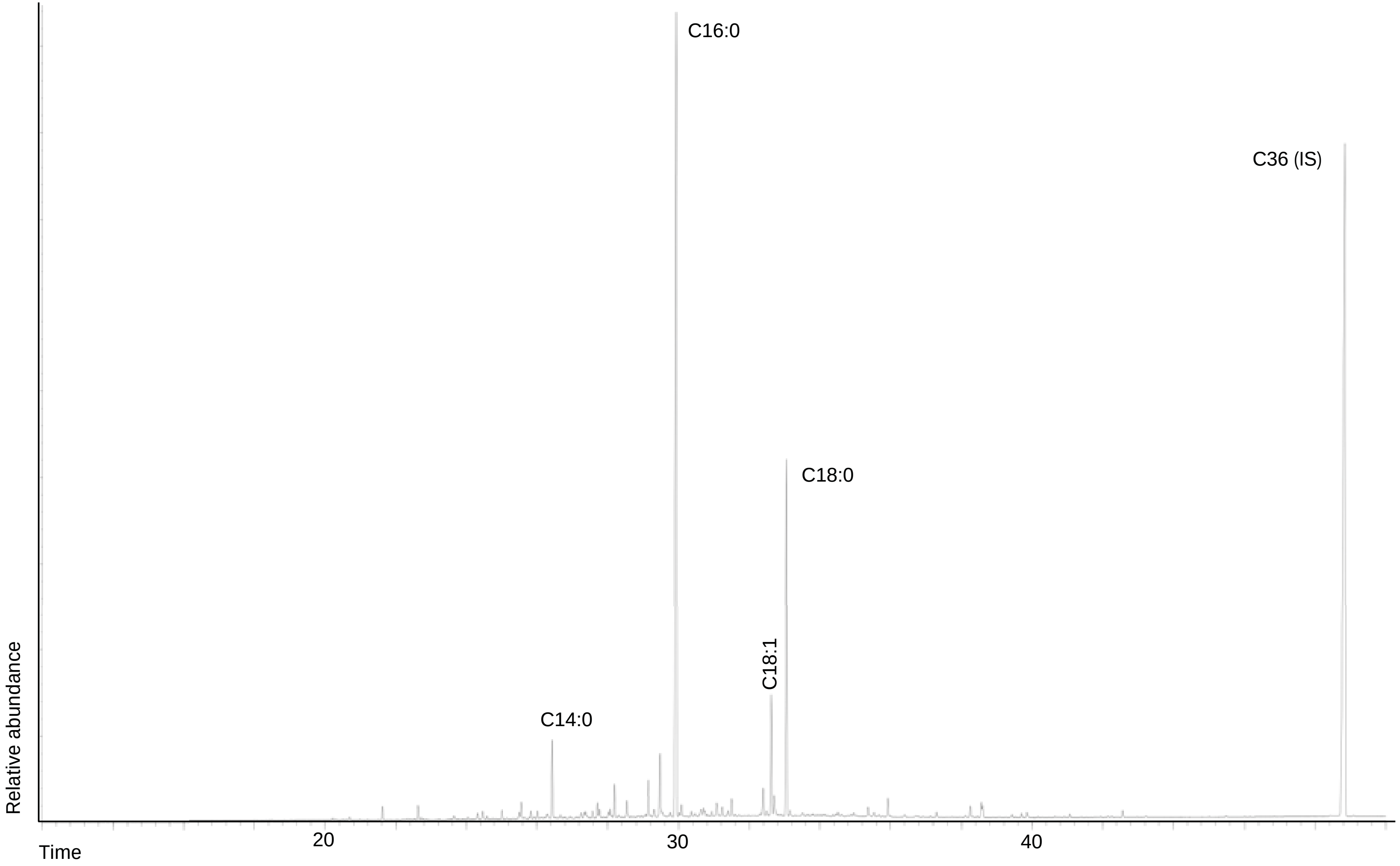

2-hydroxy fatty acids (traces):
C16:0
C18:0
C24:0
C26:0
C16:0
C18:0
C36 (IS)
C14:0
C15:0br
C15:0
C17:0br
C17:0
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
C28:0
Cholesterol
C34 (IS)
Relative abundance
Time
10
20
30

CS21

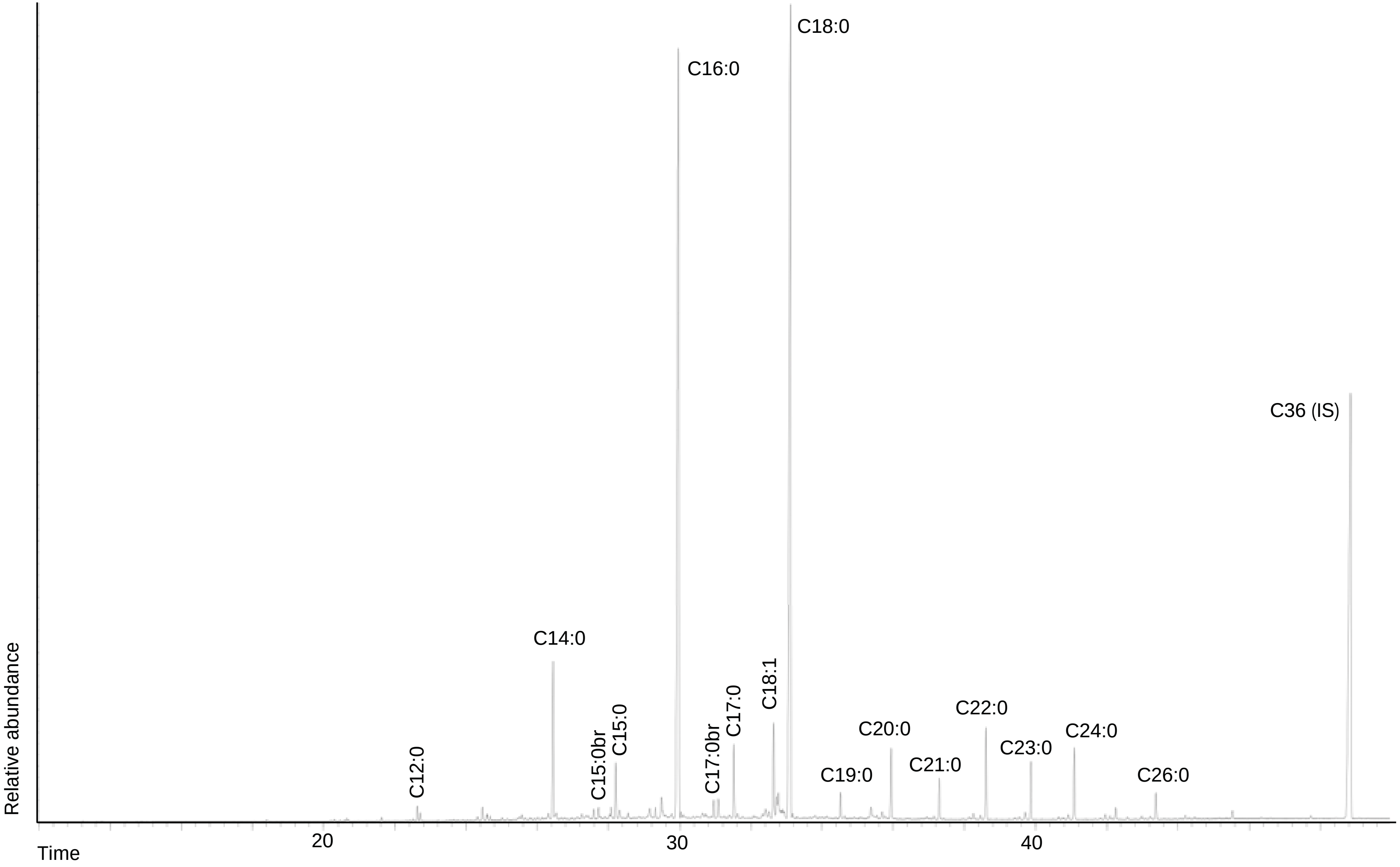

Relative abundance
Time
10
20
30
C12:0
D9
C14:1
C14:0
C15:0
C16:0
C17:0br
C17:0
phytanic acid
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
C34 (IS)
C36 (IS)

La Serreta

SE1

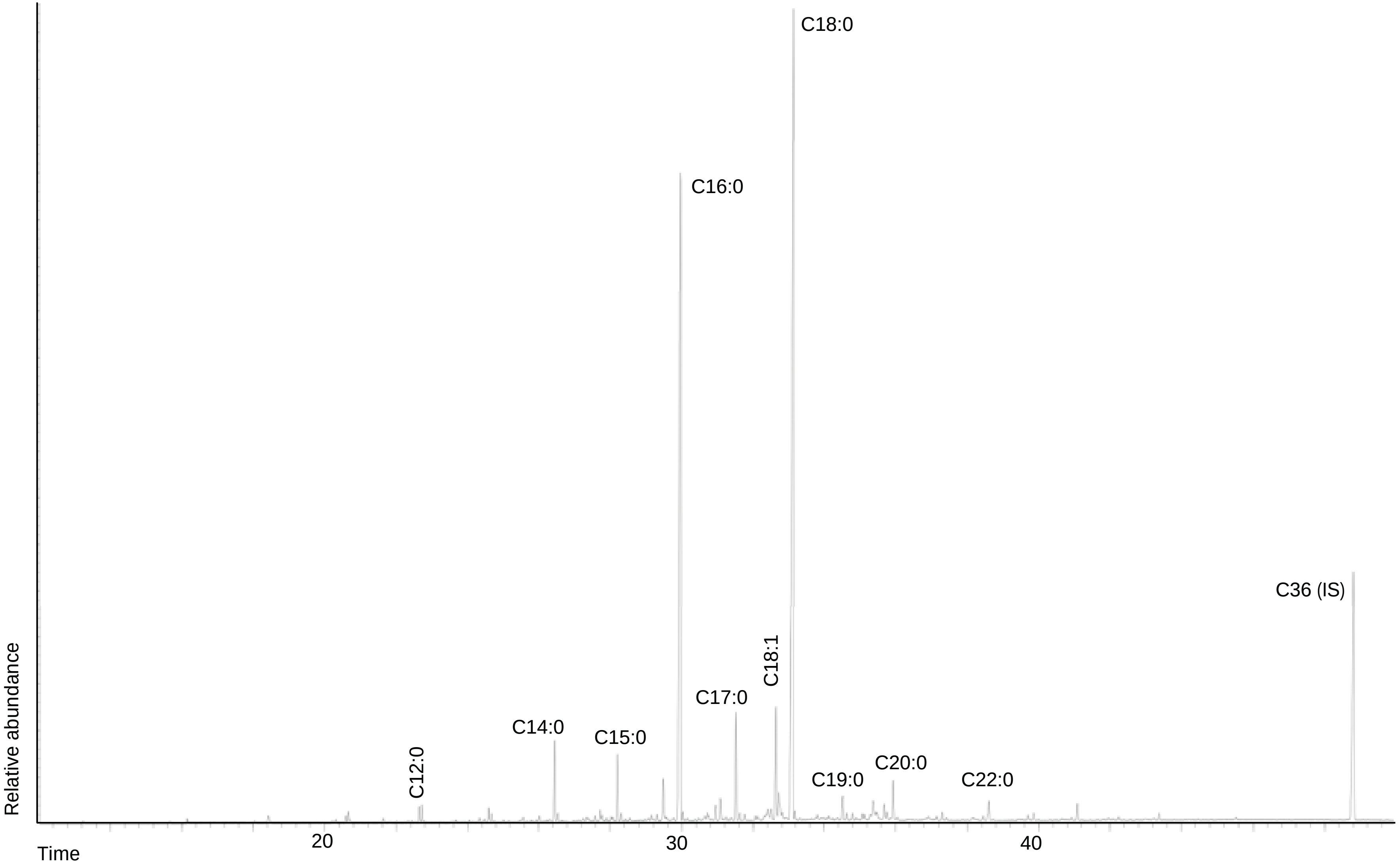


SE3

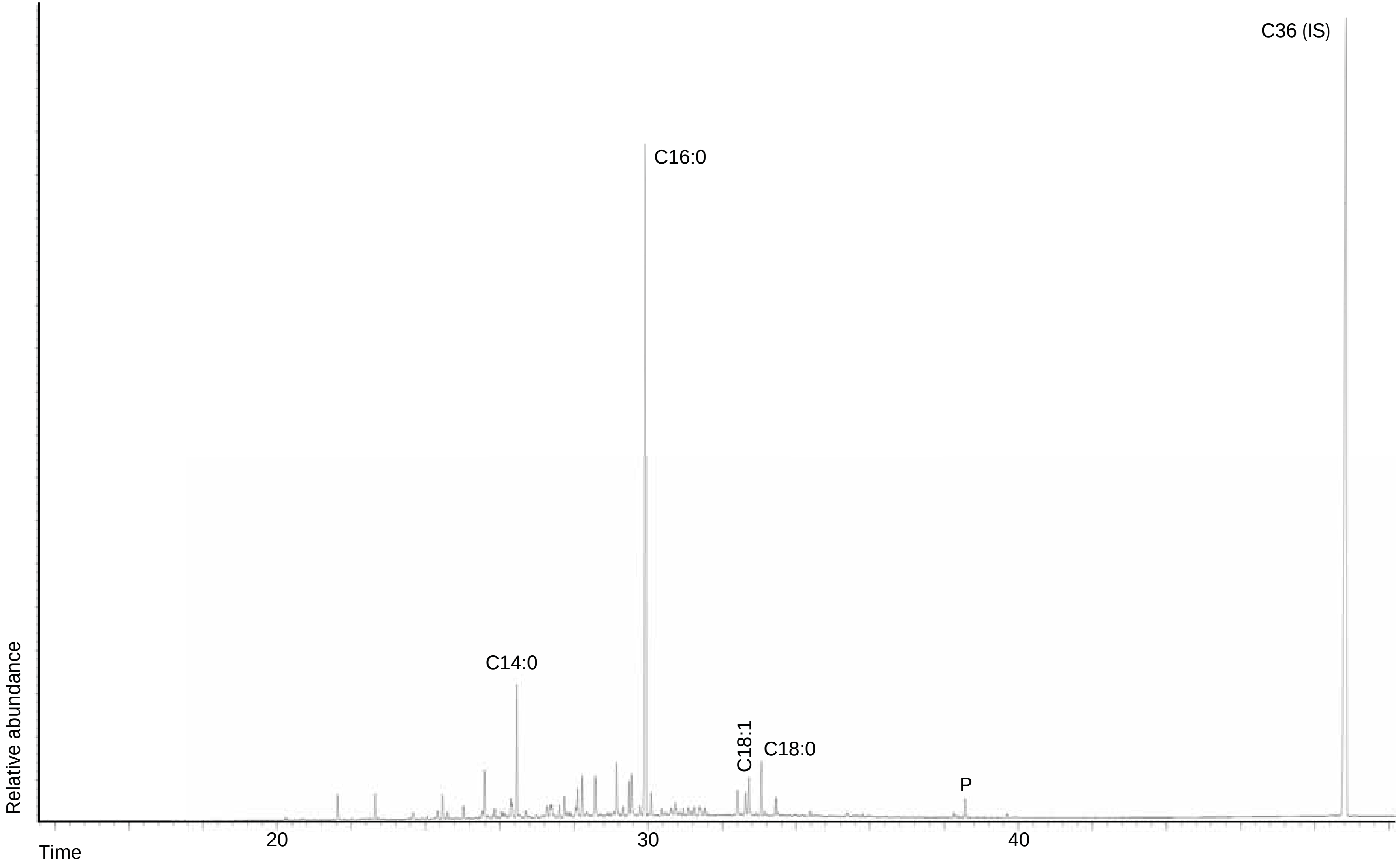


Relative abundance
C12:0
C14:0
C15:0br
C15:0
C16:0
C17:0br
C17:0
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C26:0
C36 (IS)
Time
20
30
40

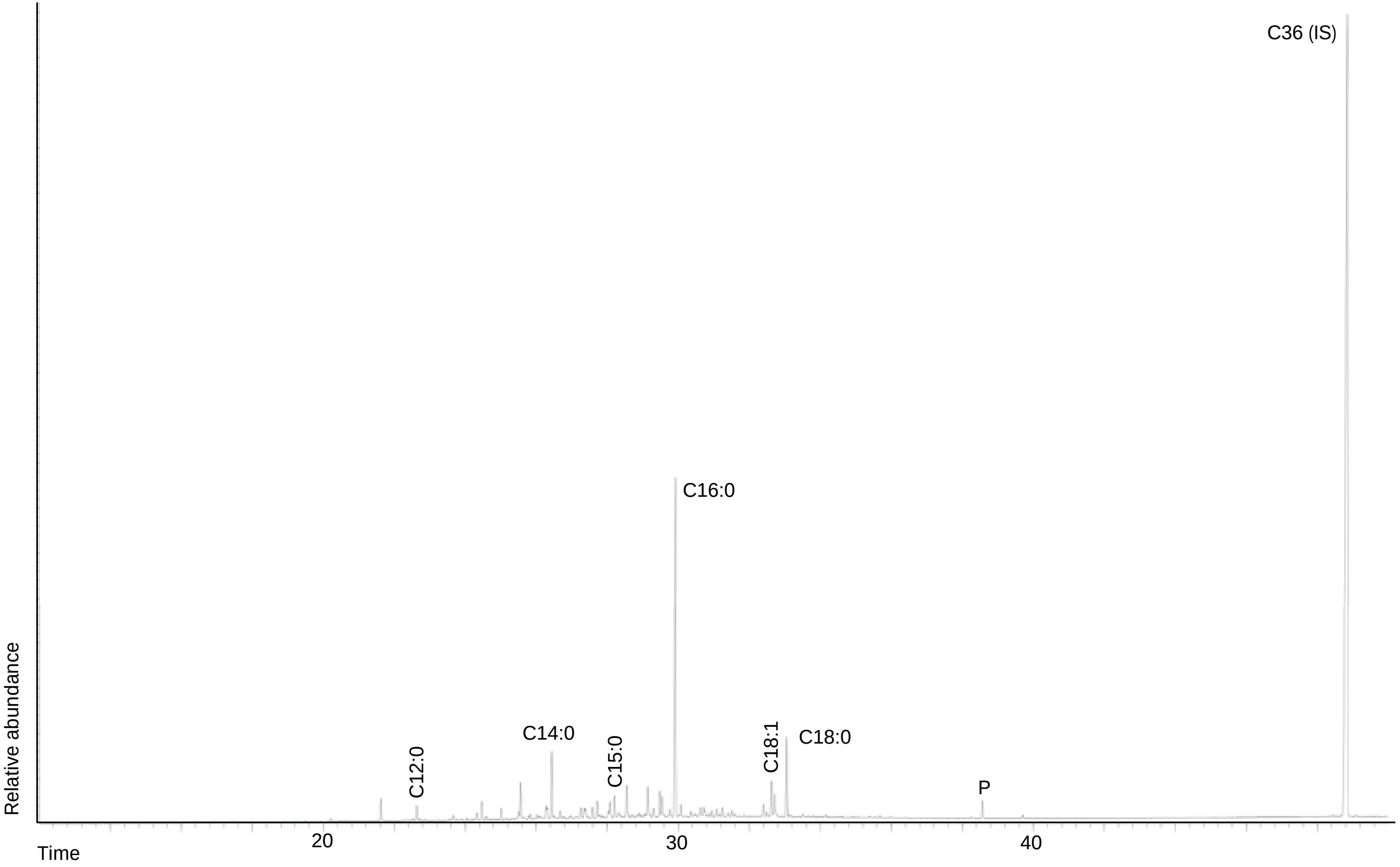
SE5


C18:0
C16:0
C36 (IS)
C18:1
C17:0
C14:0
C15:0
C12:0
C19:0
C20:0
C22:0
Relative abundance
Time
20
30
40

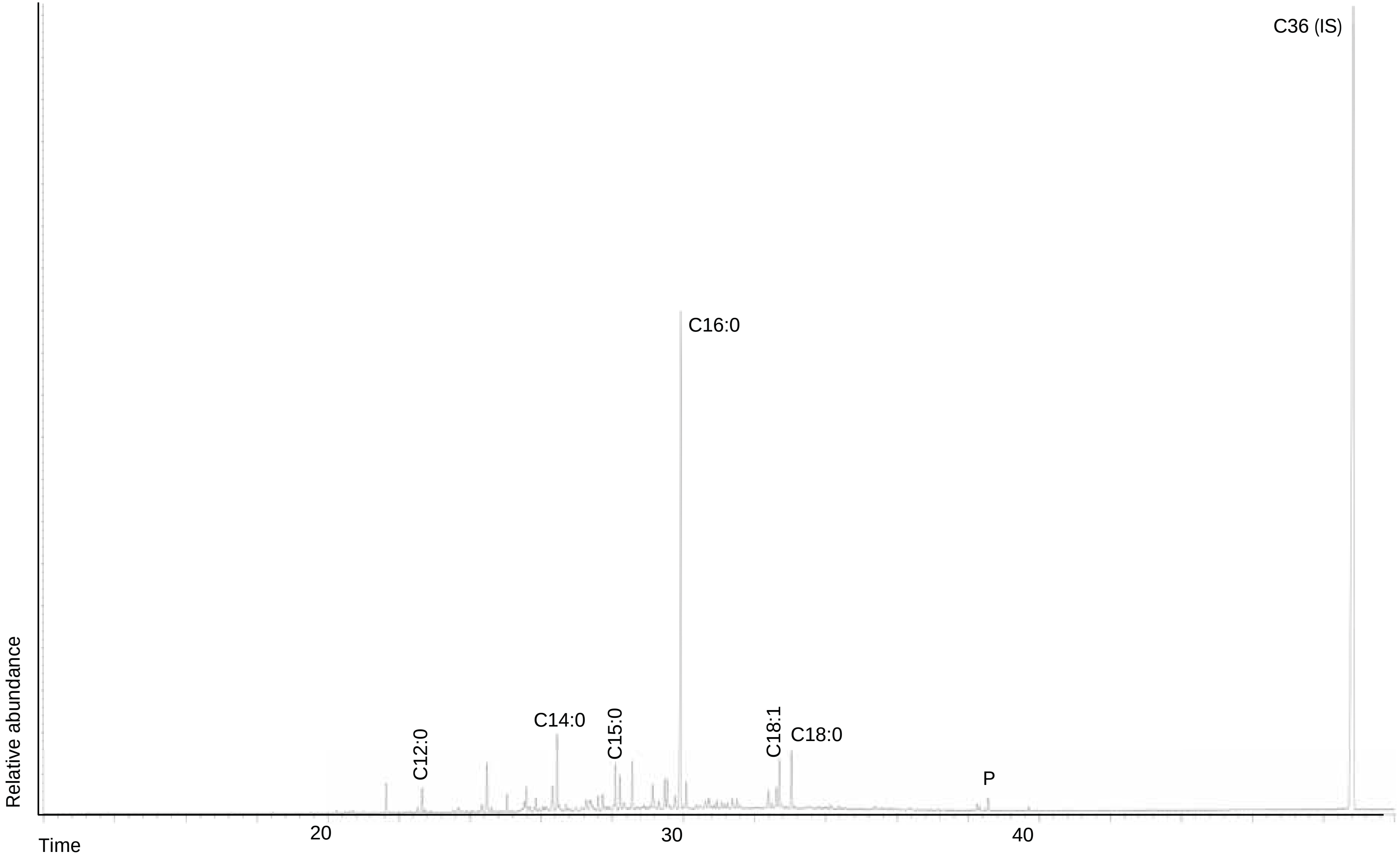
SE9



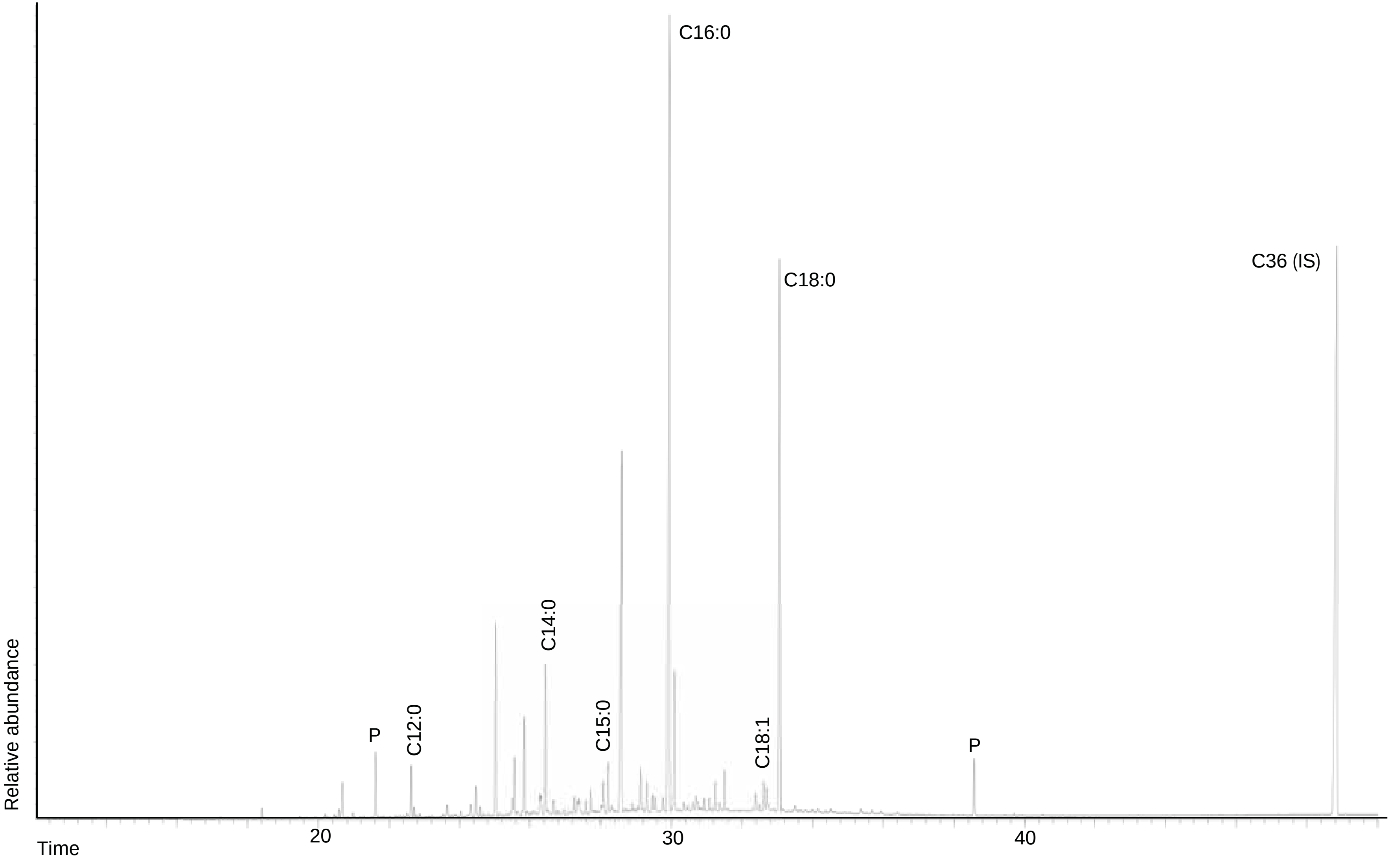
SE10


C36 (IS)
C16:0
C14:0
C12:0
C15:0
C18:1
C18:0
P
Relative abundance
Time
20
30
40

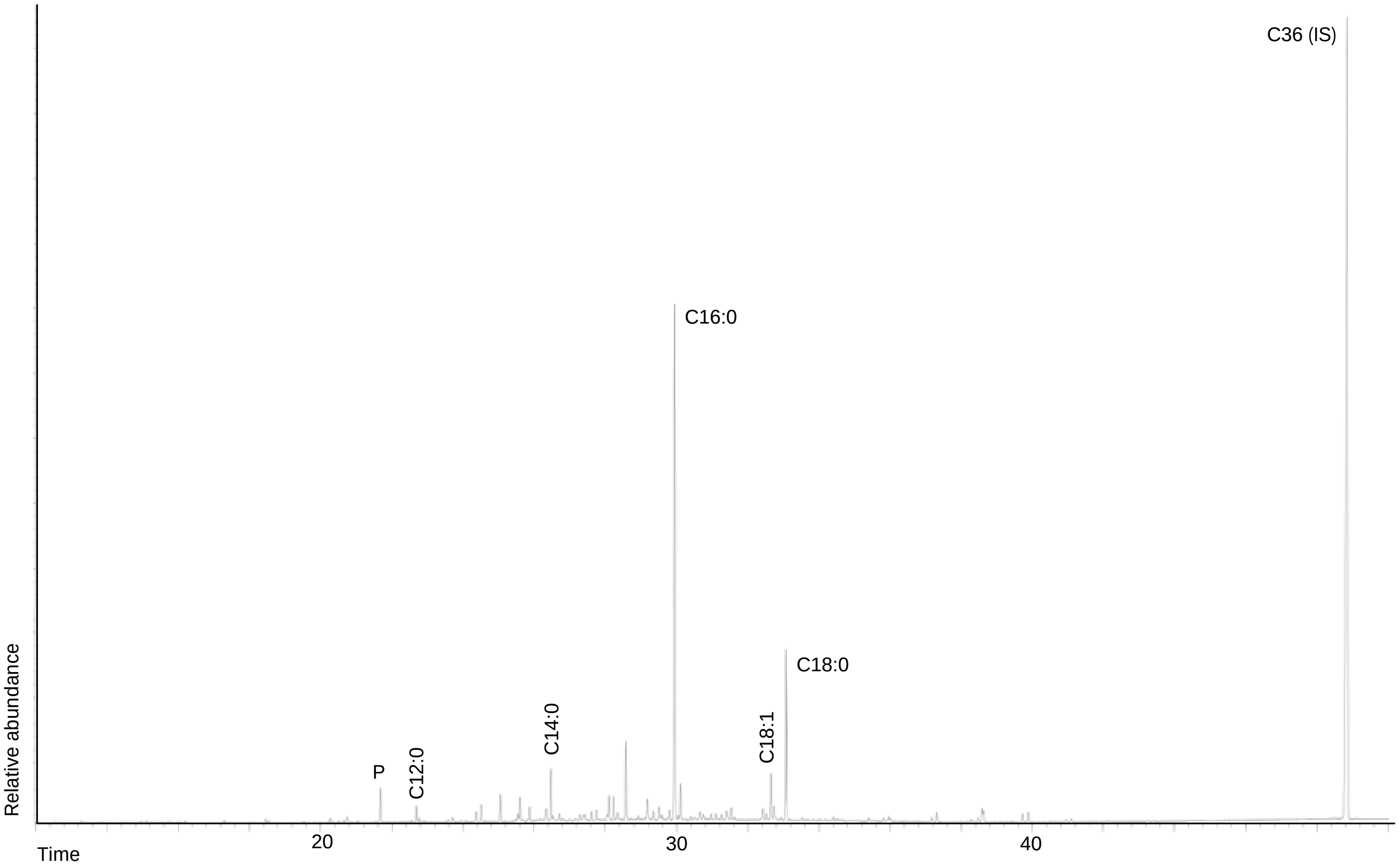
SE12


C36 (IS)
C16:0
C12:0
C14:0
C15:0
C18:1
C18:0
P
Relative abundance
Time
20
30
40

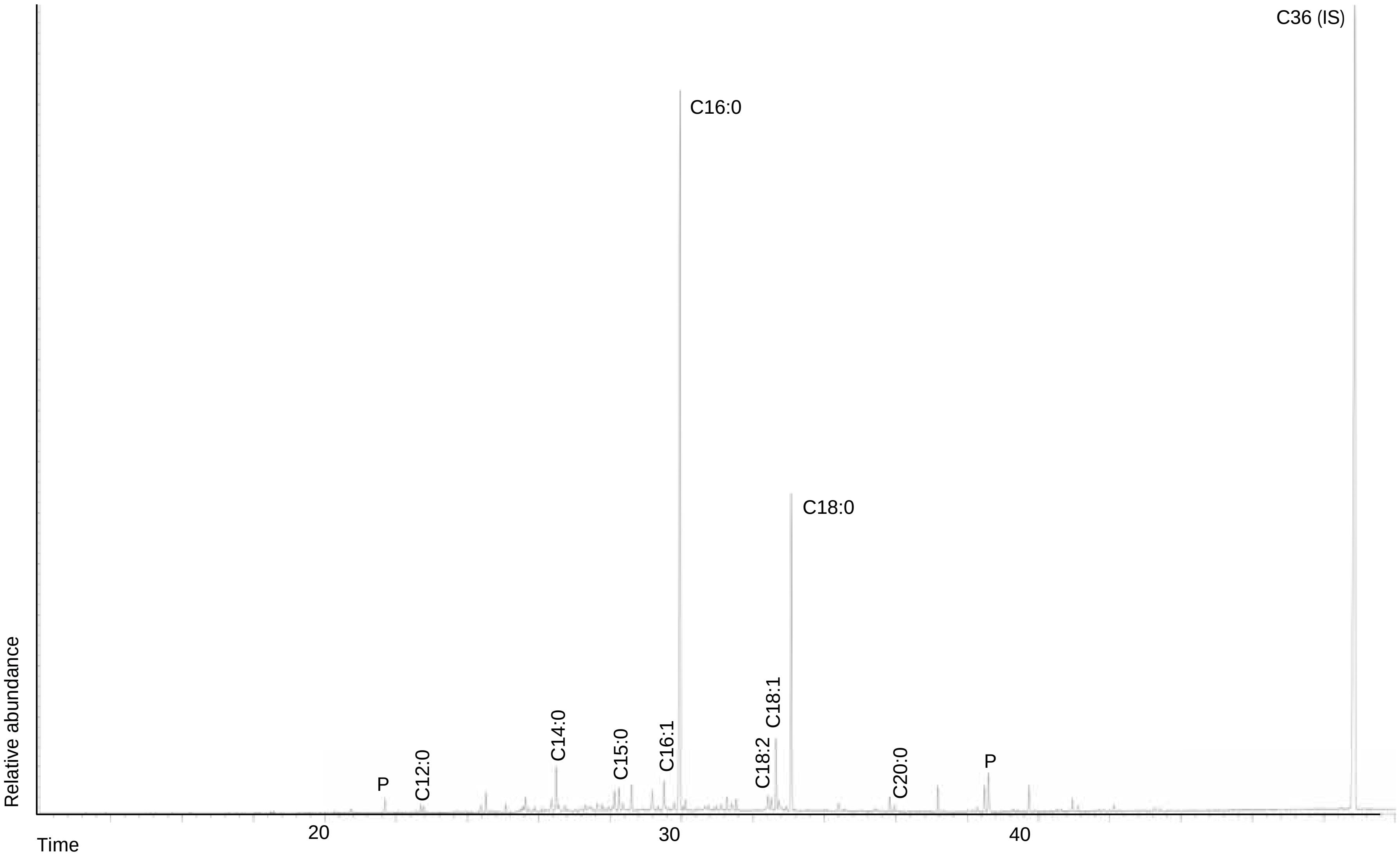
SE13


C16:0
C18:0
C36 (IS)
C14:0
P
C12:0
C15:0
C18:1
P
Relative abundance
Time
20
30
40

SEP1

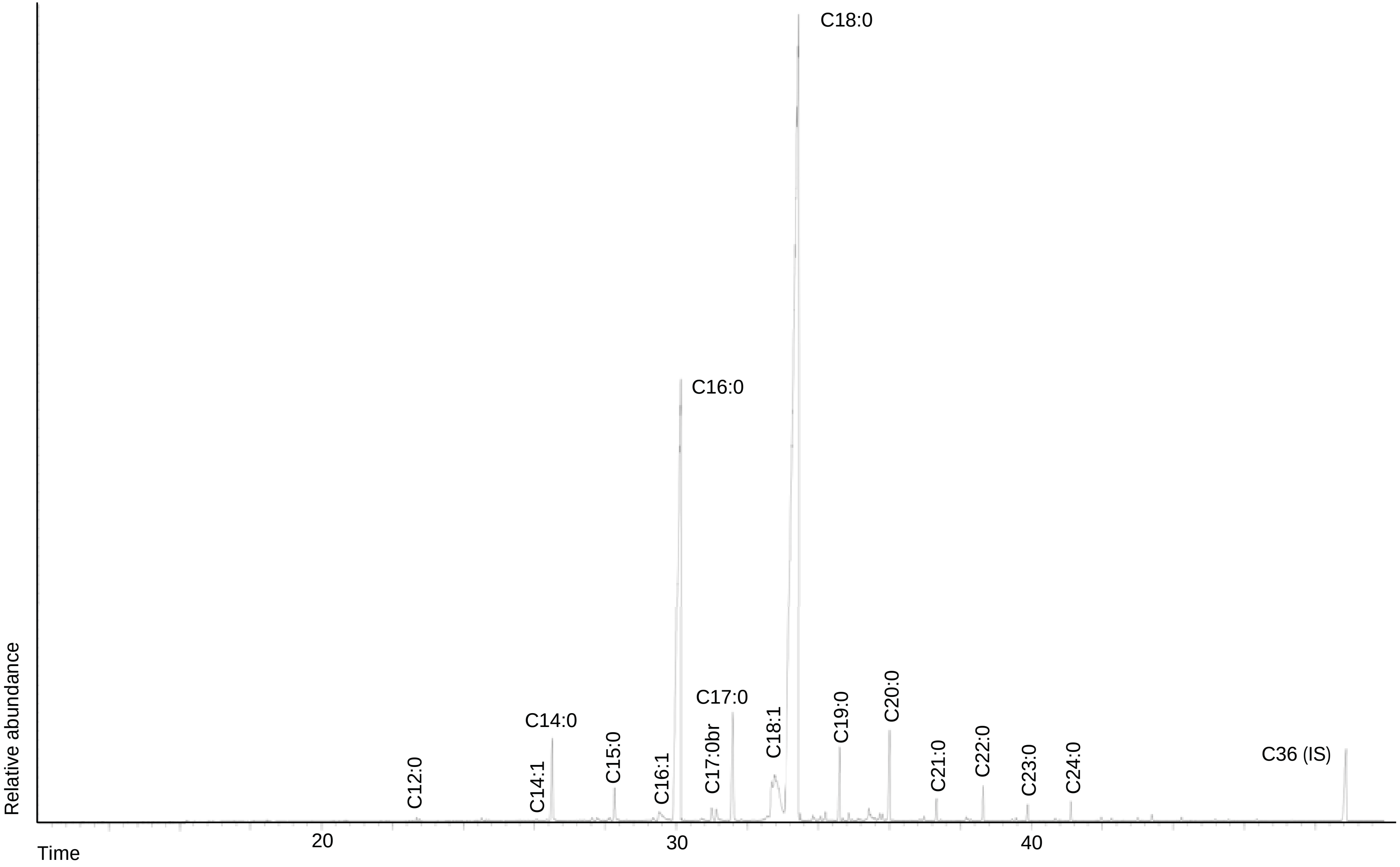

C36 (IS)
C16:0
C18:0
C14:0
C18:1
P
C12:0
Relative abundance
Time
20
30
40

SEP2

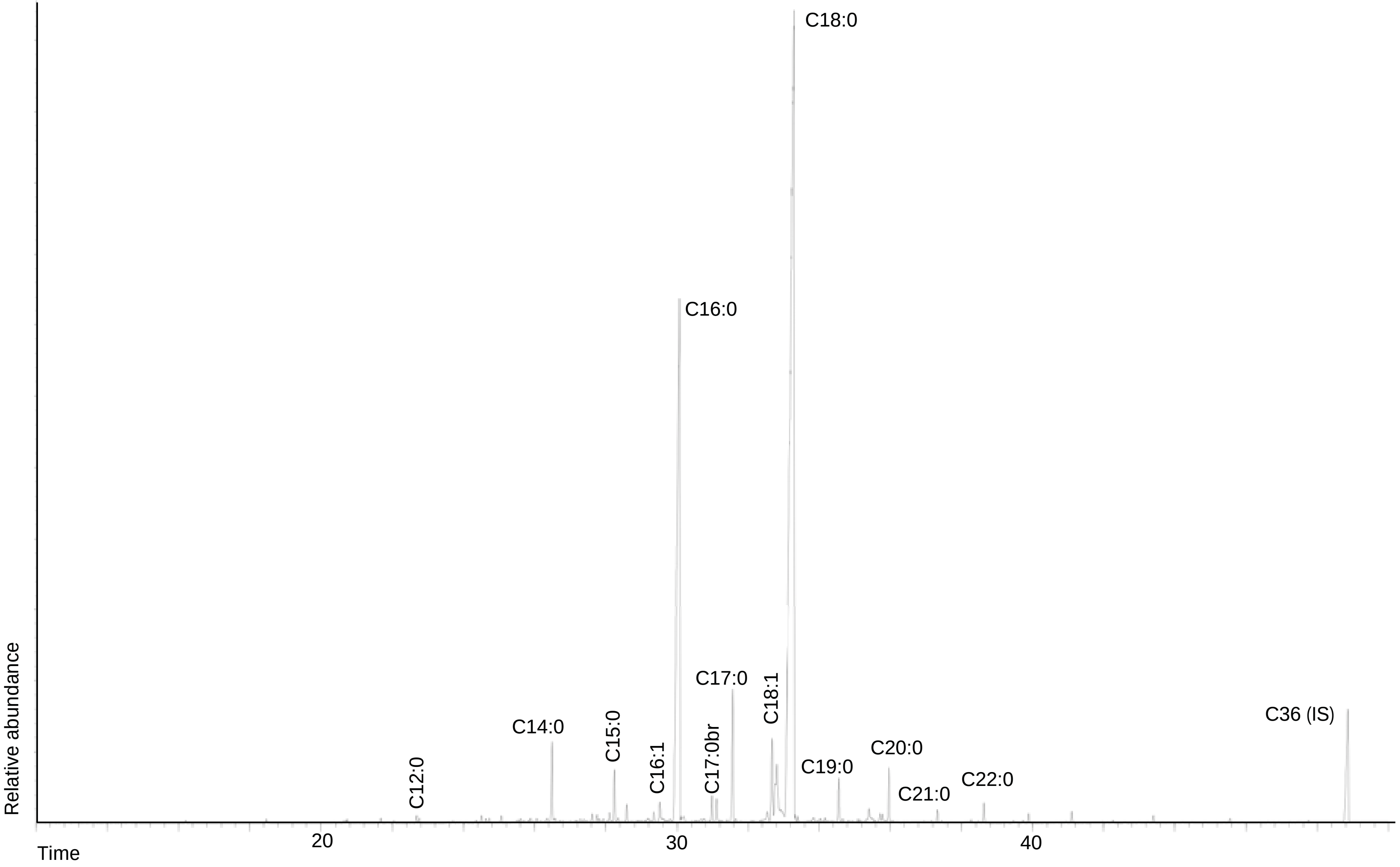


C36 (IS)
C16:0
C18:0
C18:1
C14:0
C15:0
C16:1
C18:2
C20:0
P
C12:0
P
Relative abundance
Time
20
30
40

SEP4

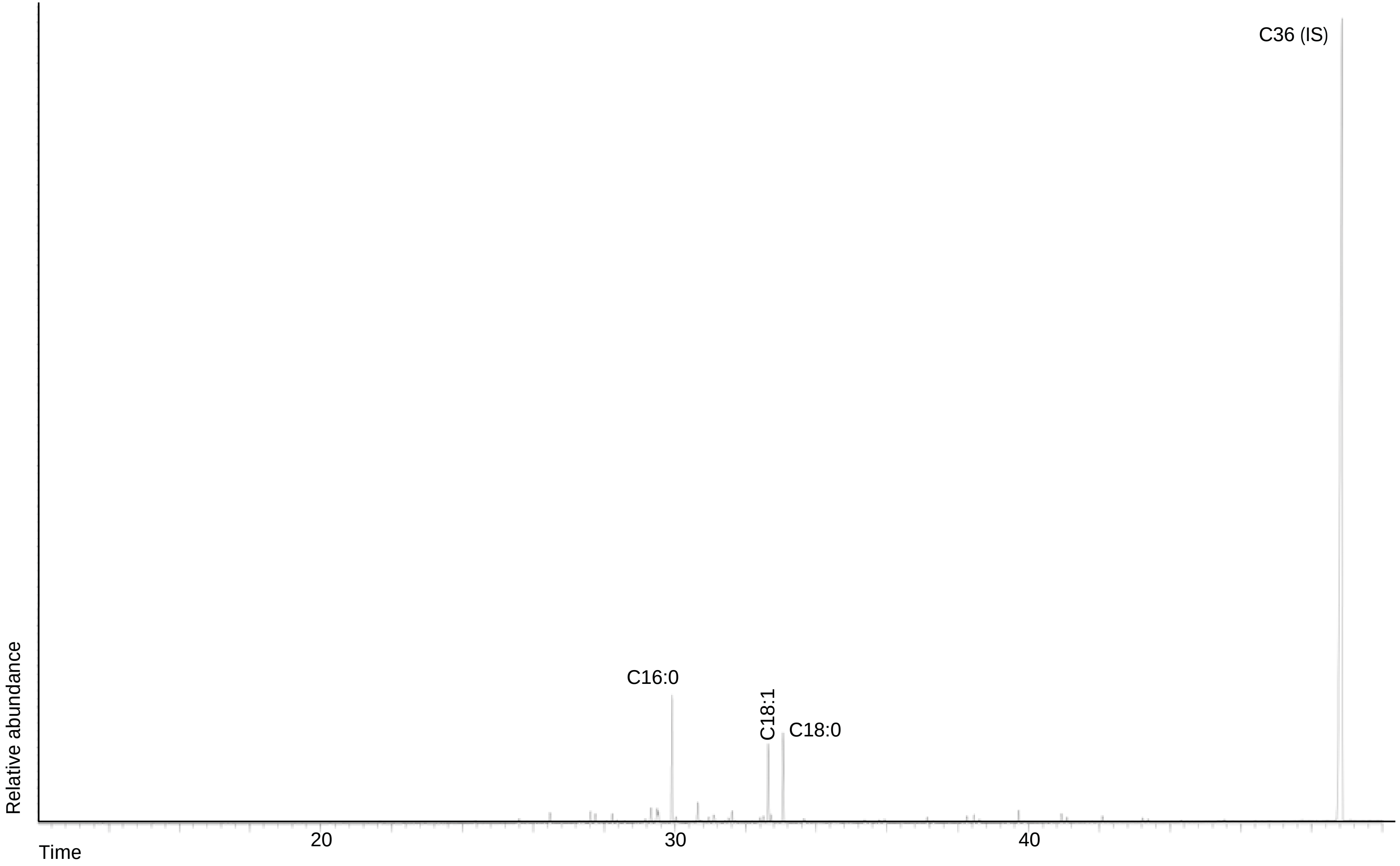

C18:0
C16:0
C17:0
C14:0
C20:0
C19:0
C18:1
C17:0br
C16:1
C15:0
C14:1
C12:0
C21:0
C22:0
C23:0
C24:0
C36 (IS)
Relative abundance
Time
20
30
40

SEP5

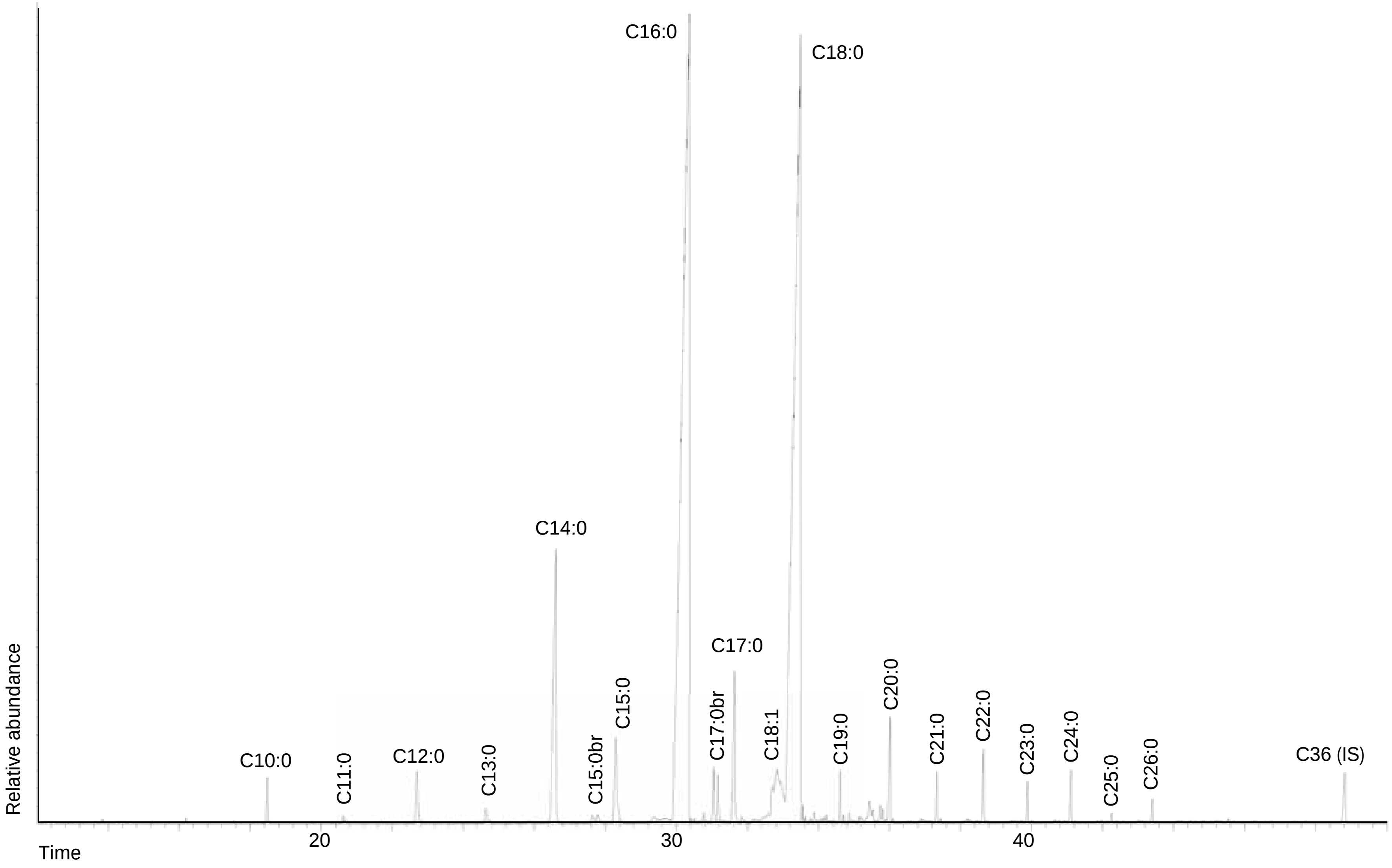

C18:0
C16:0
C17:0
C18:1
C14:0
C15:0
C16:1
C17:0br
C12:0
C19:0
C20:0
C21:0
C22:0
C36 (IS)
Relative abundance
Time
20
30
40

Guixeres de Vilobí

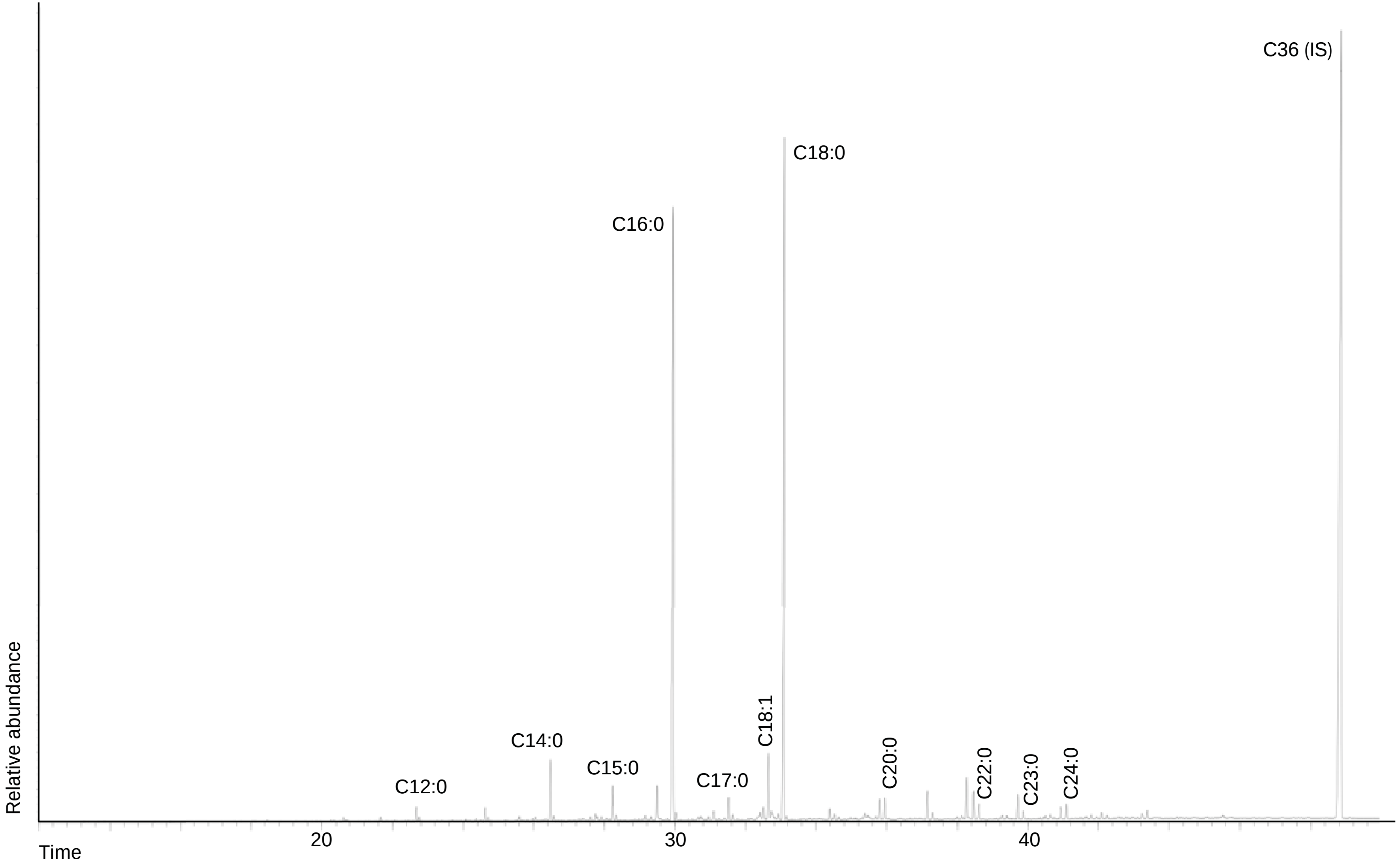
GV20



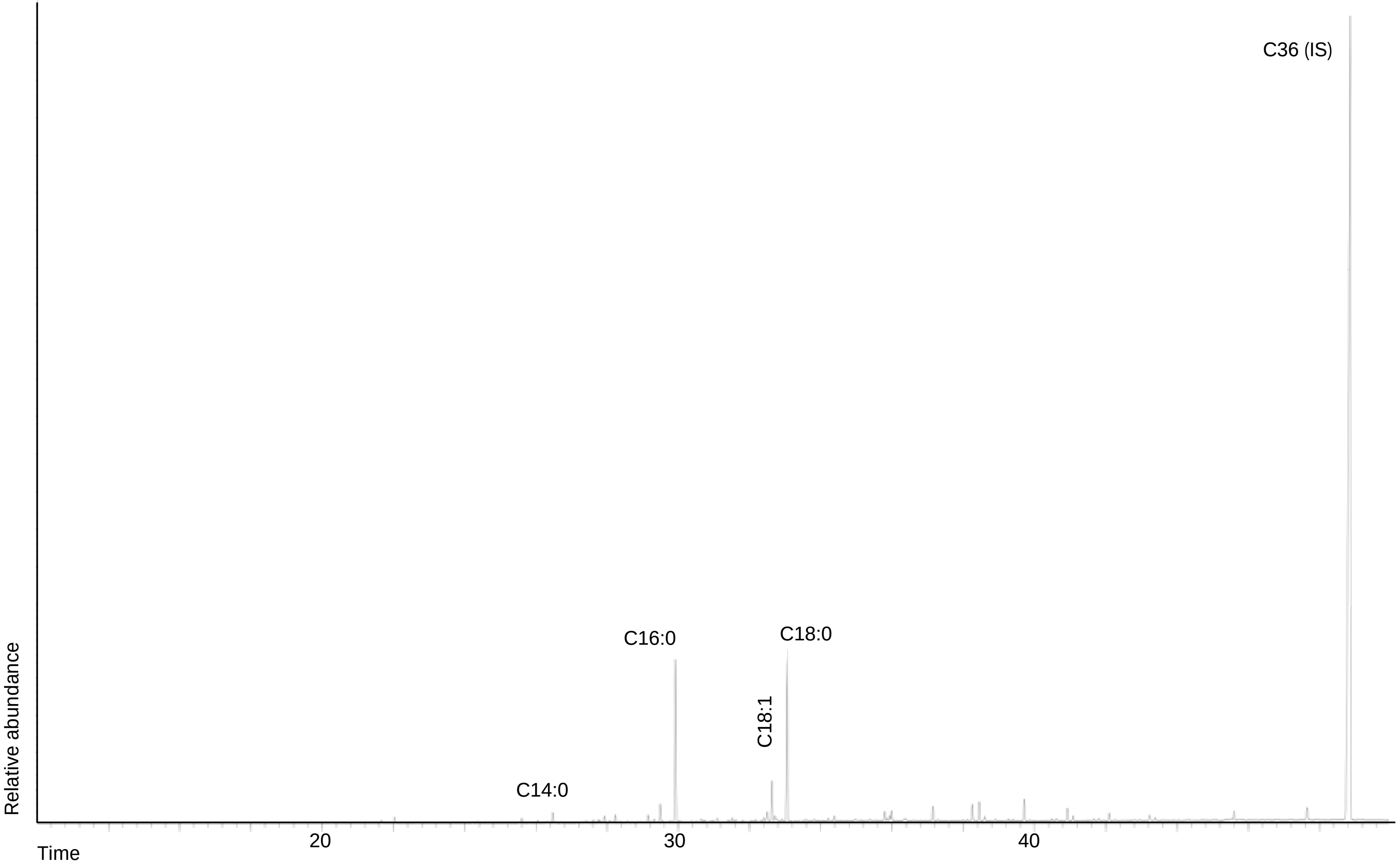
GV23


C16:0
C18:0
C14:0
C17:0
C10:0
C11:0
C12:0
C13:0
C15:0br
C15:0
C17:0br
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
C36 (IS)
Relative abundance
Time
20
30
40

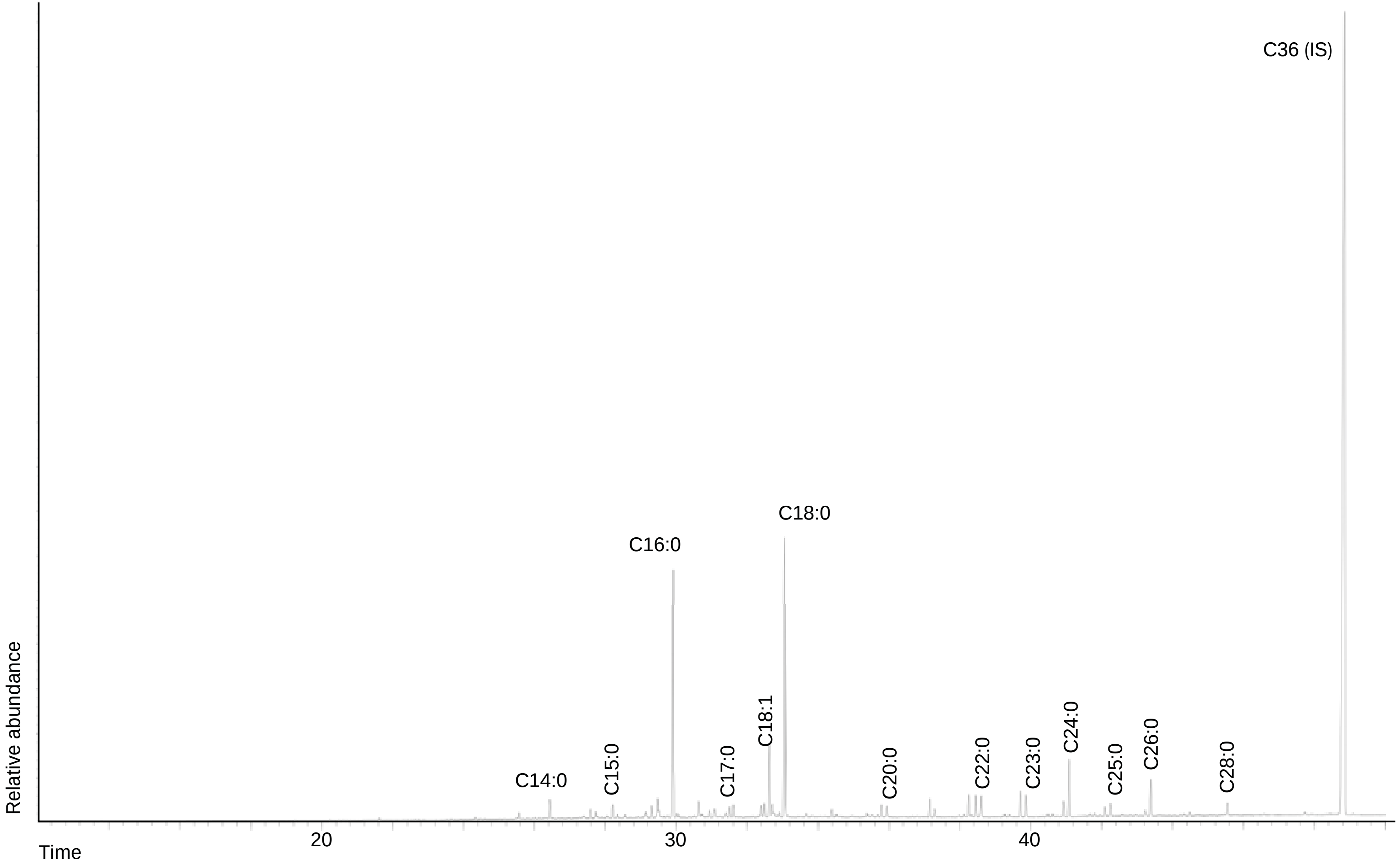
GV28F


C36 (IS)
C18:0
C16:0
C18:1
C14:0
C15:0
C12:0
C17:0
C20:0
C22:0
C23:0
C24:0
Relative abundance
Time
20
30
40

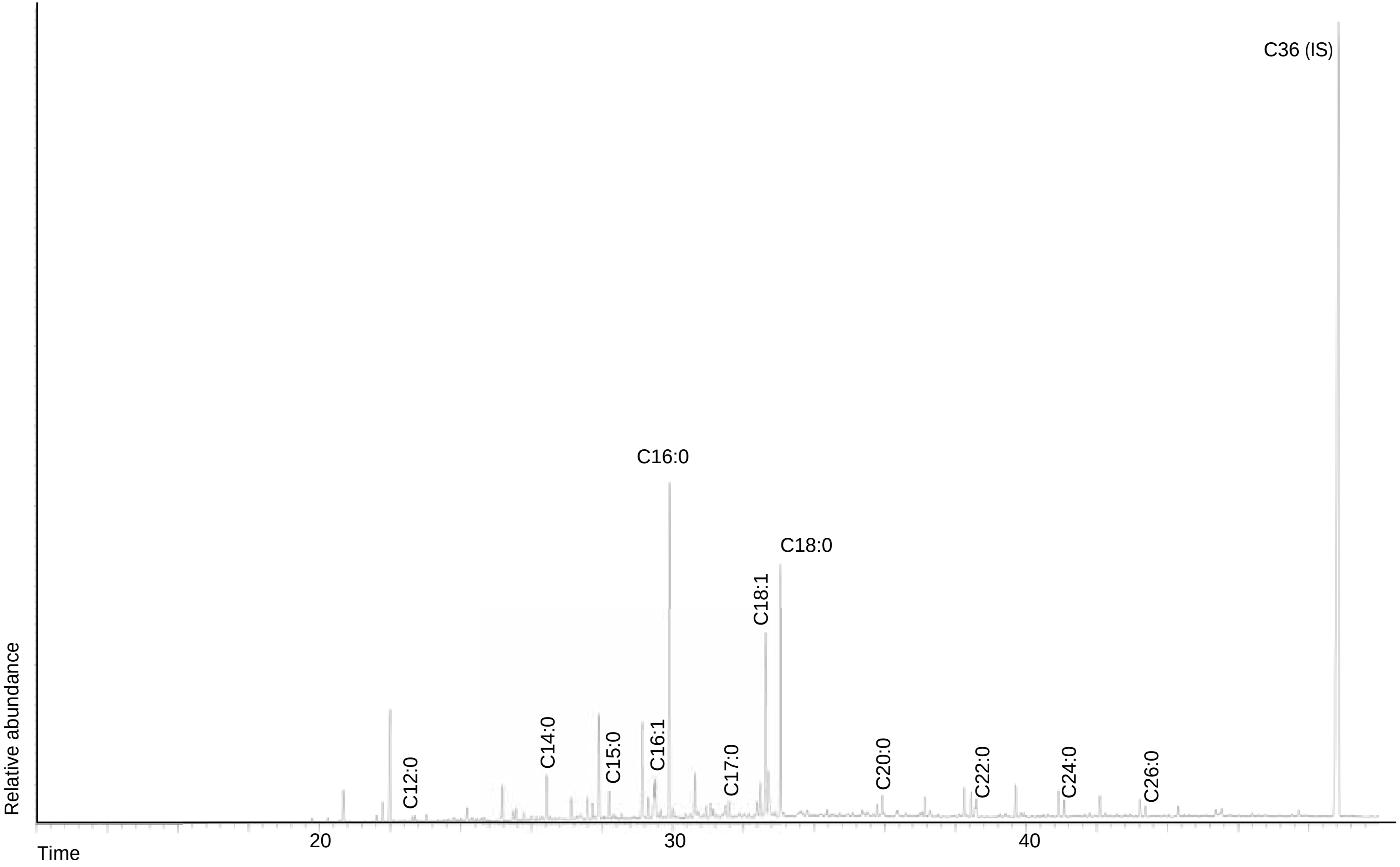
GV28K



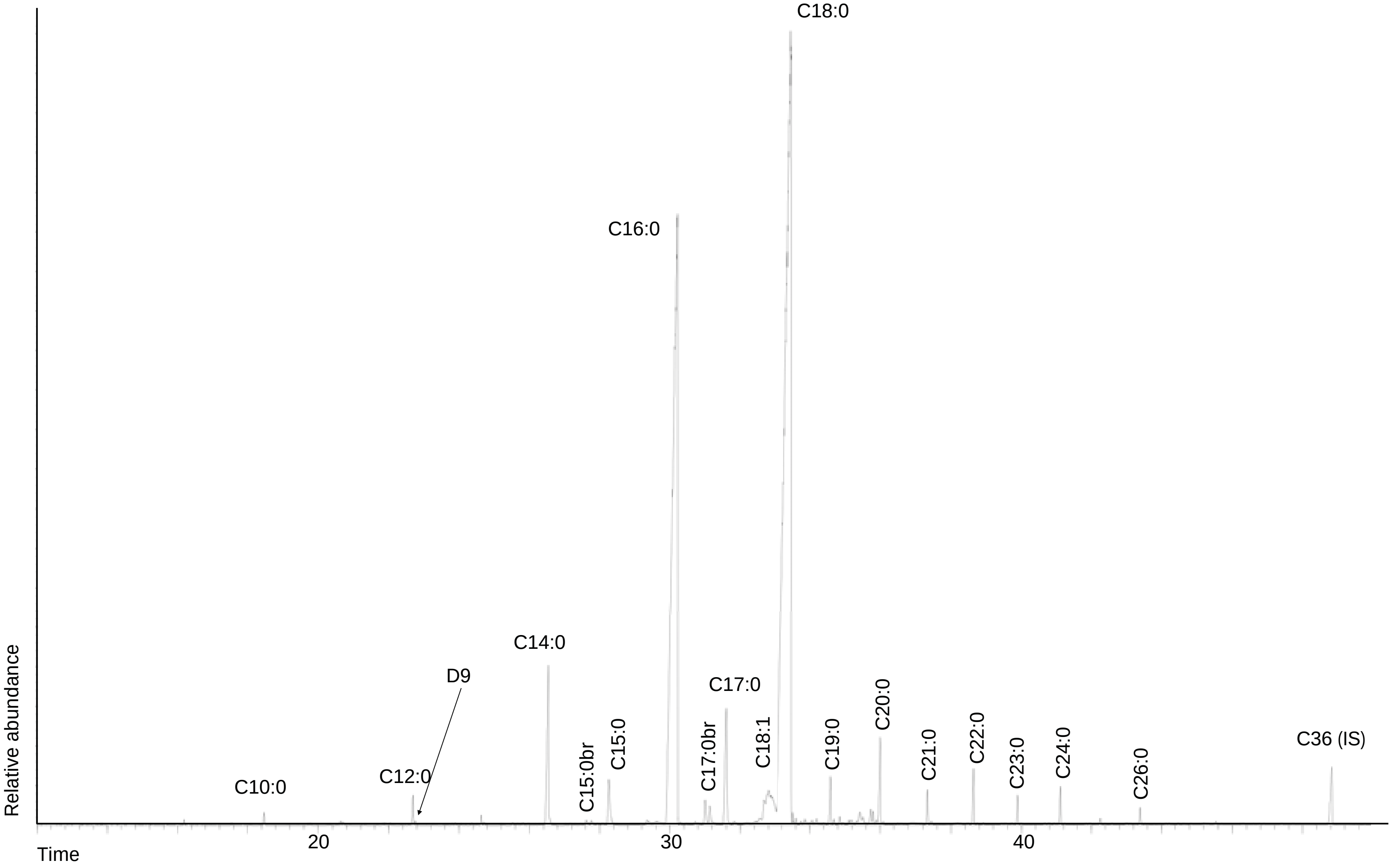
GV42


C36 (IS)
C18:0
C16:0
C18:1
C24:0
C26:0
C14:0
C15:0
C17:0
C20:0
C22:0
C23:0
C25:0
C28:0
Relative abundance
Time
20
30
40

GV43

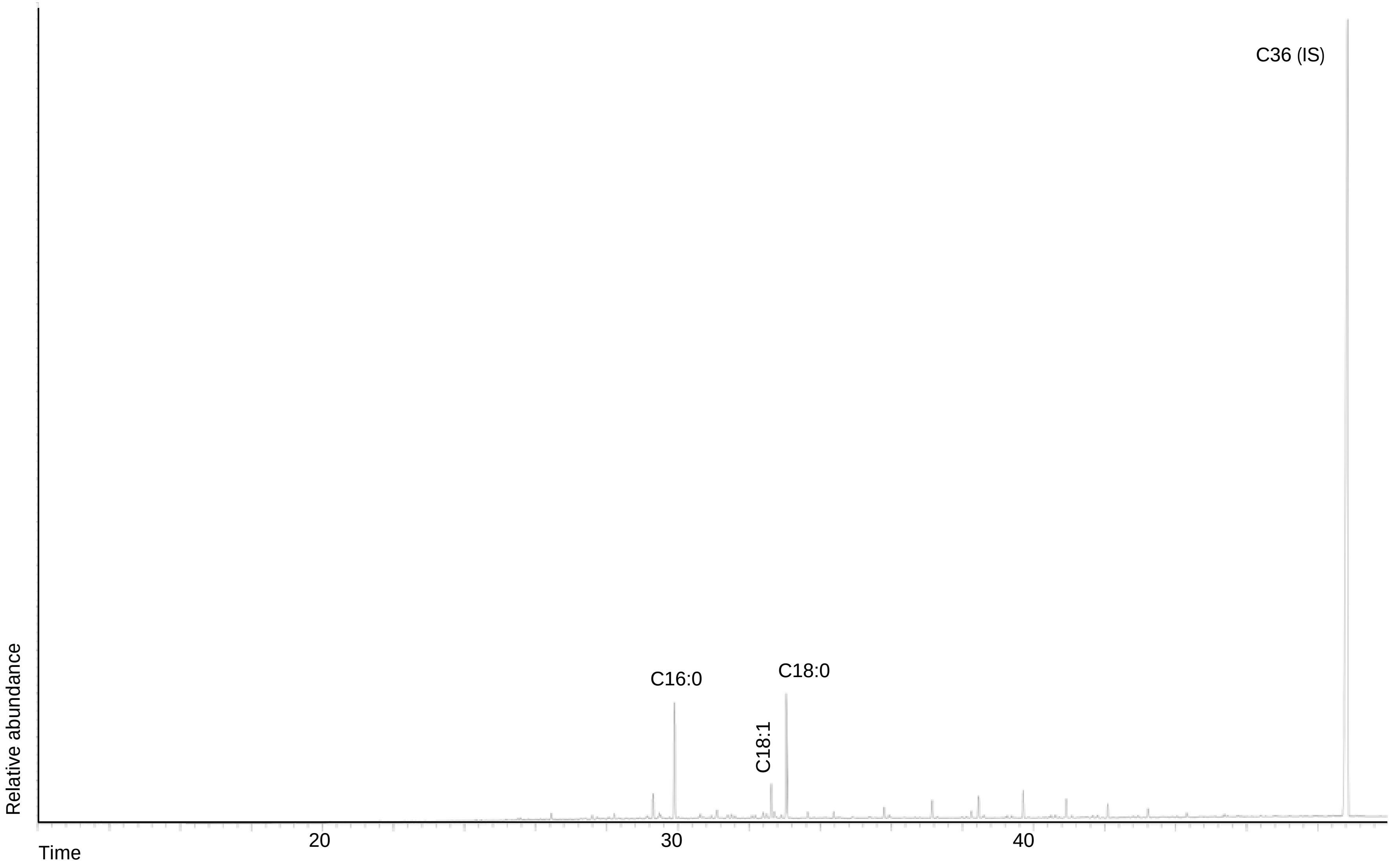

C36 (IS)
C16:0
C18:0
C18:1
C12:0
C14:0
C15:0
C16:1
C17:0
C20:0
C22:0
C24:0
C26:0
Relative abundance
Time
20
30
40

GV44

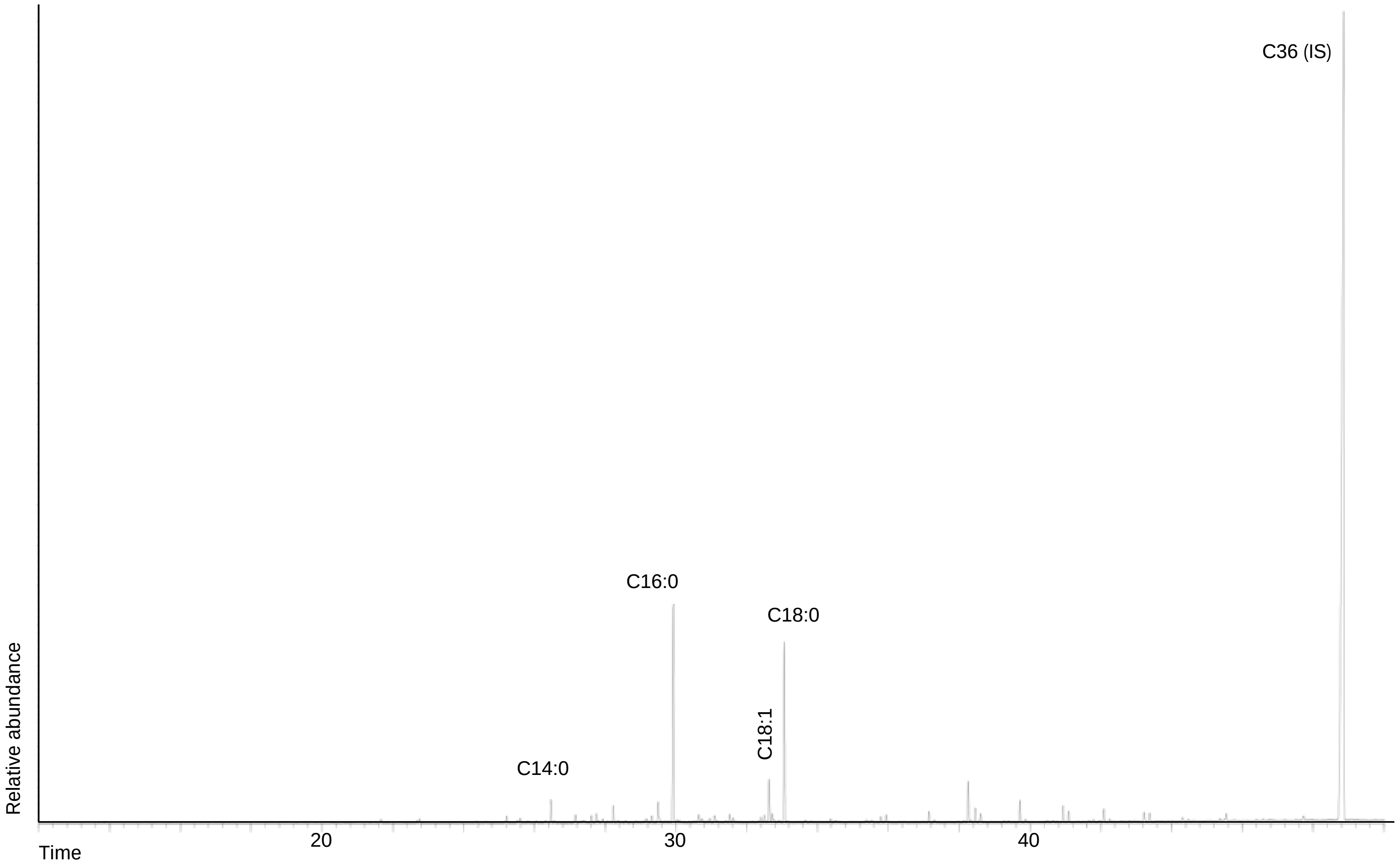


C18:0
C16:0
C14:0
D9
C17:0
C12:0
C10:0
C15:0br
C15:0
C17:0br
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C26:0
C36 (IS)
Relative abundance
Time
20
30
40

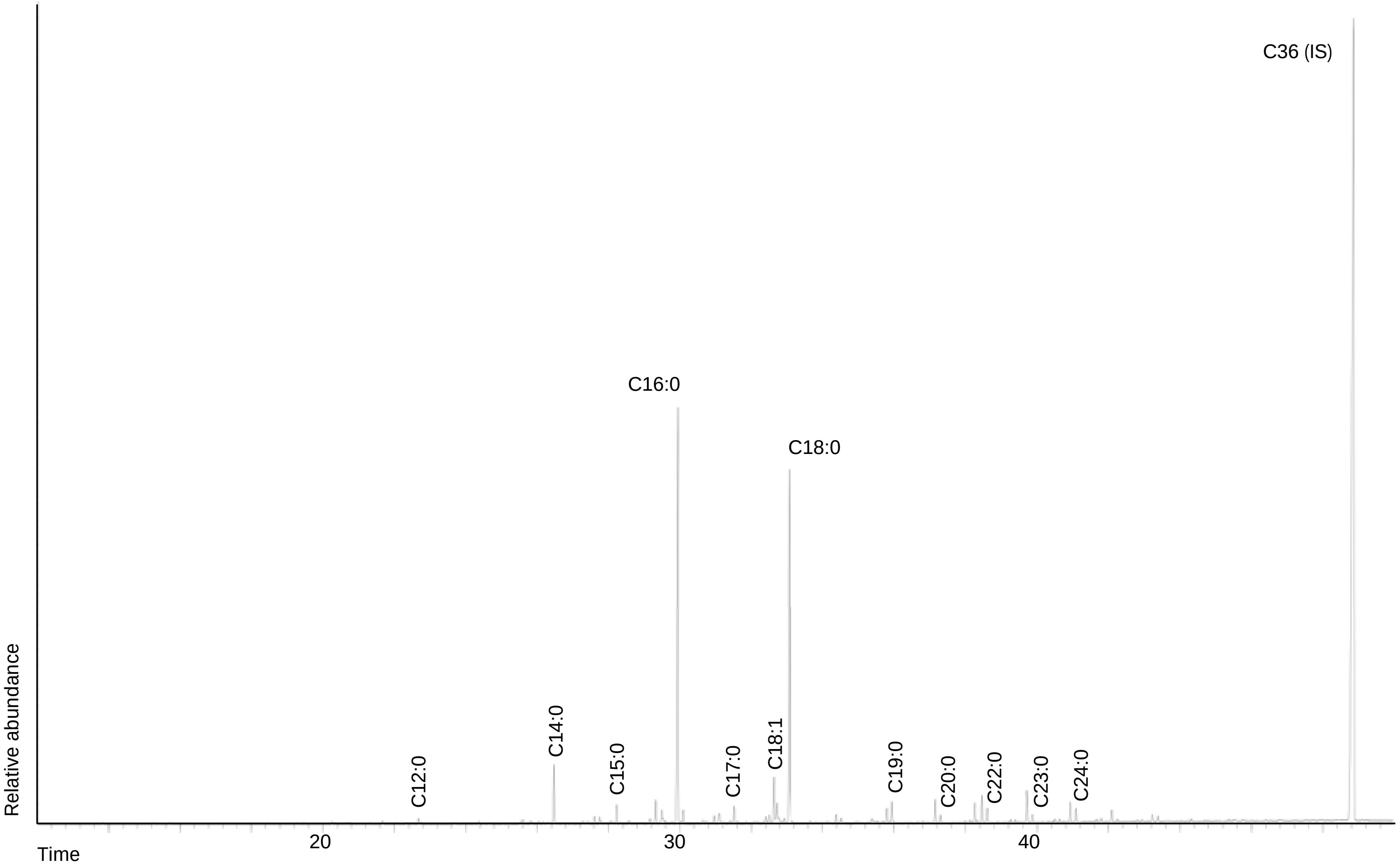
GV48



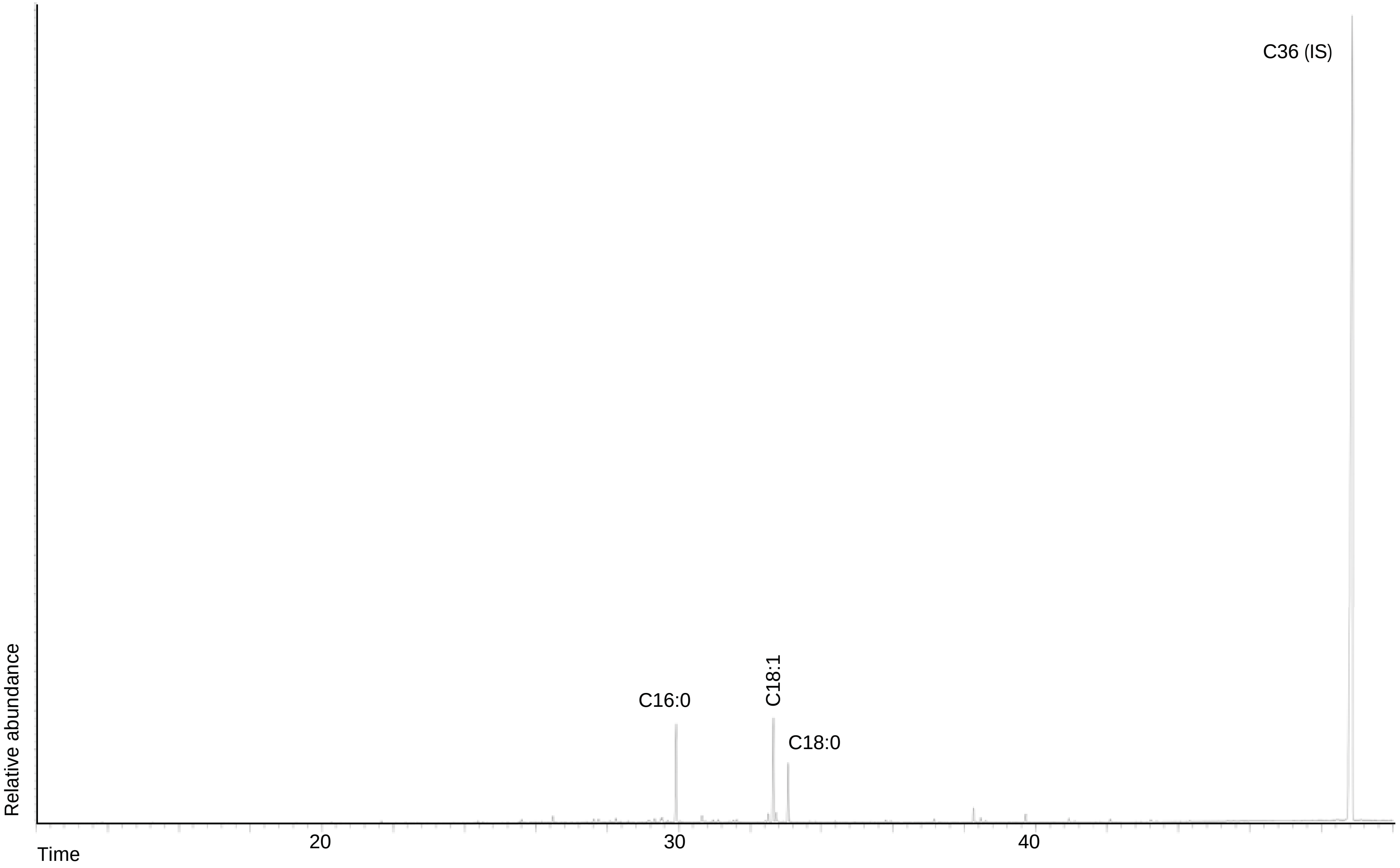
GV58



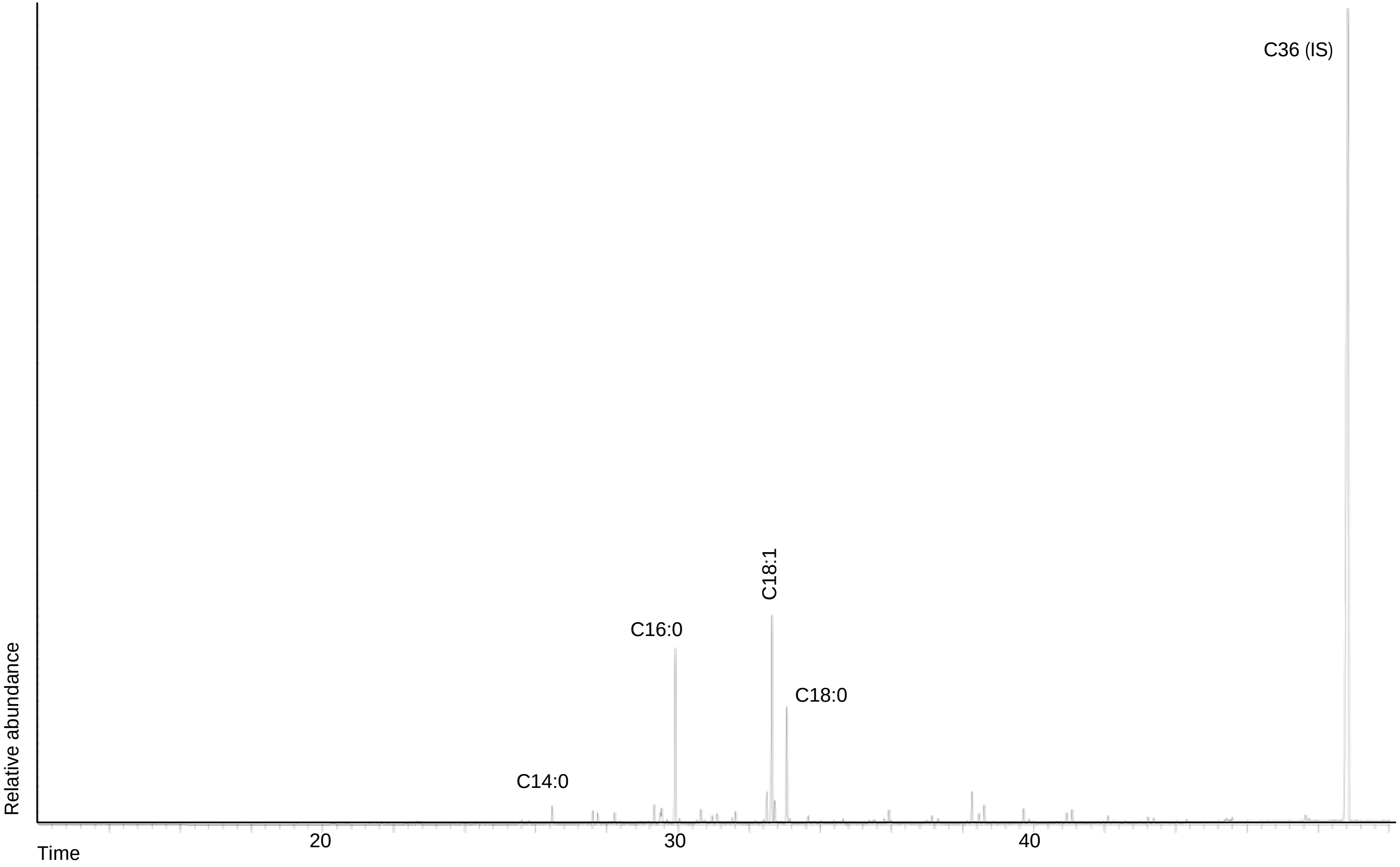
GV68


Relative abundance
C36 (IS)
C16:0
C18:0
C12:0
C14:0
C15:0
C17:0
C18:1
C19:0
C20:0
C22:0
C23:0
C24:0
Time
20
30
40

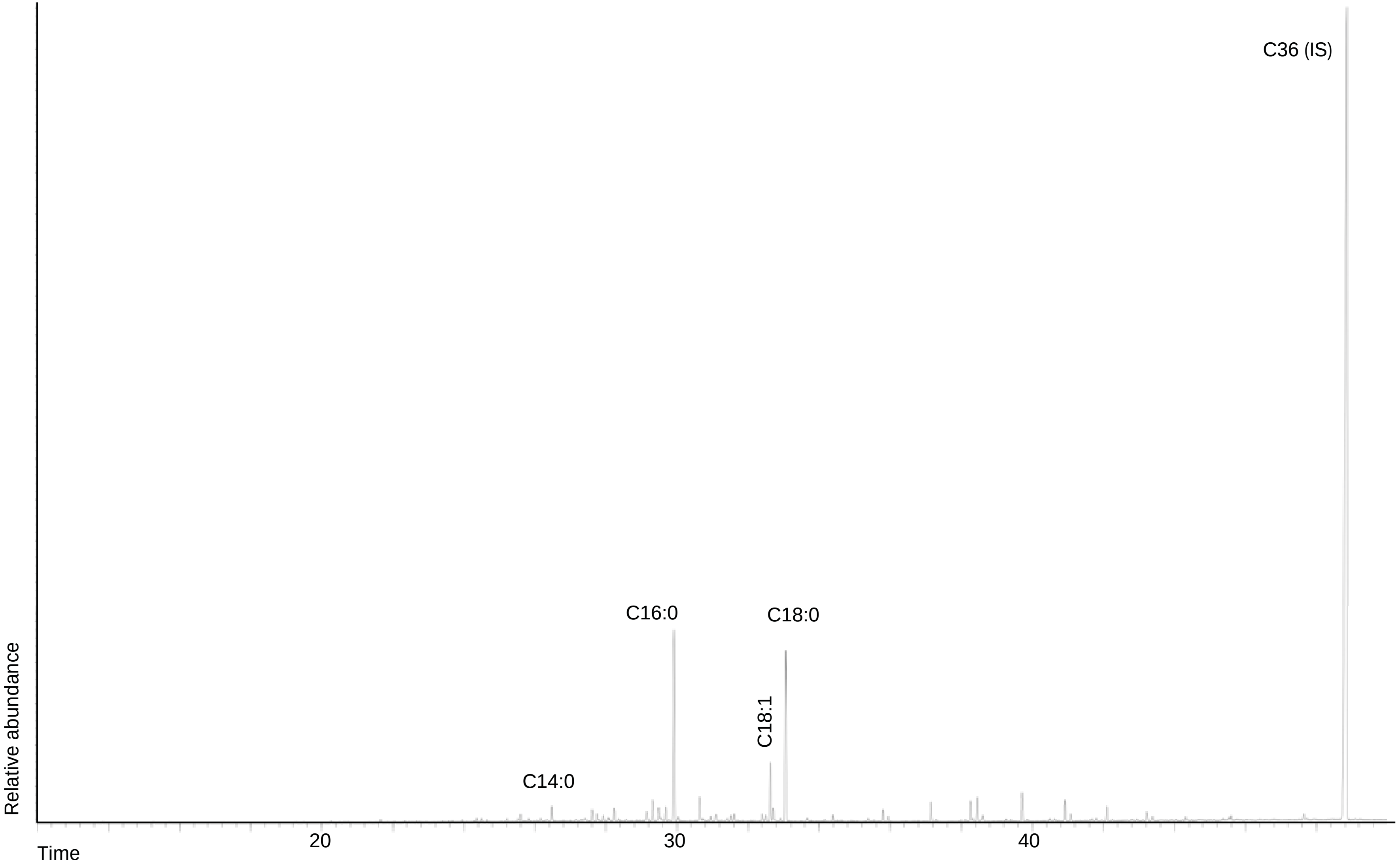
GV78



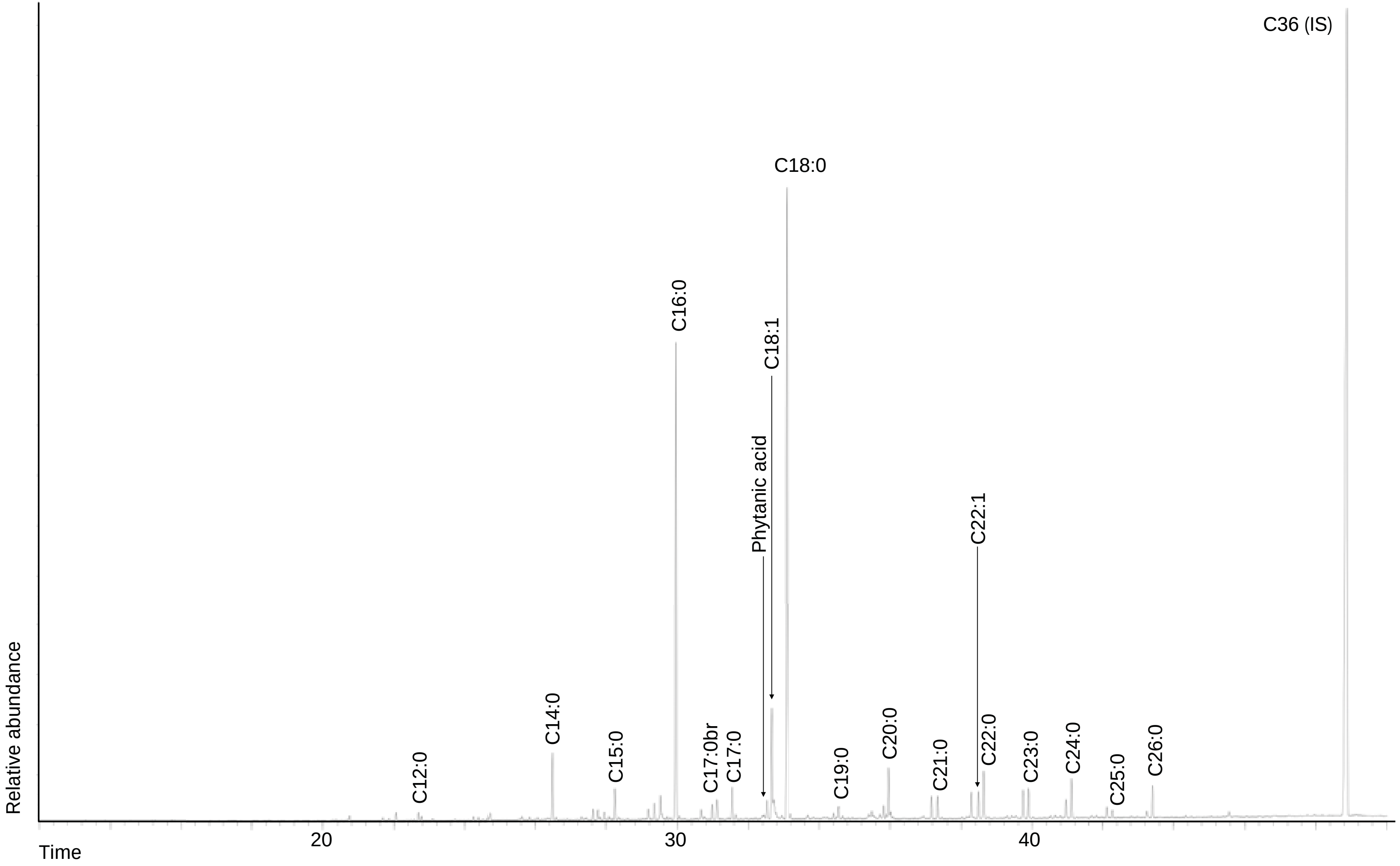
GV98



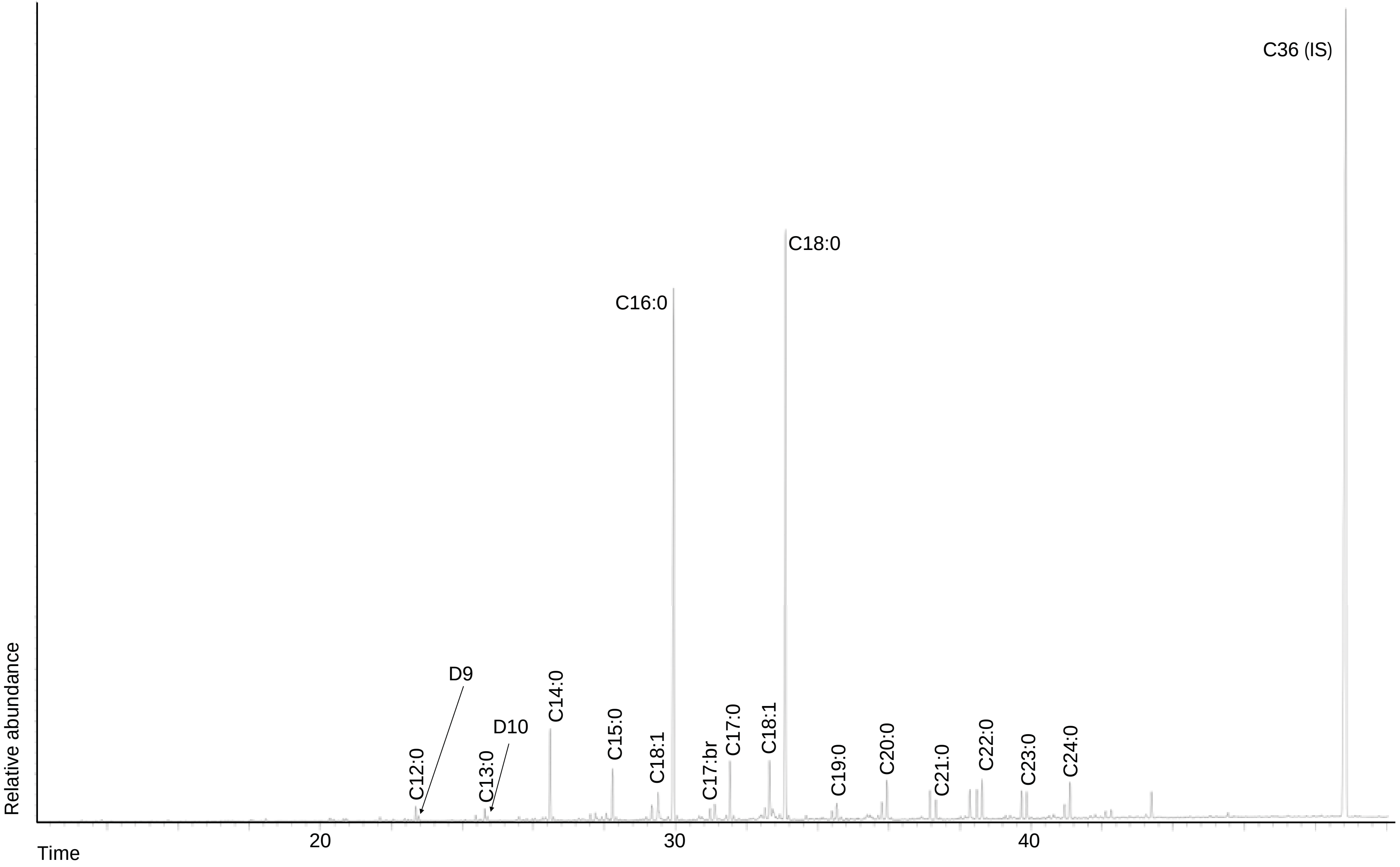
GV161



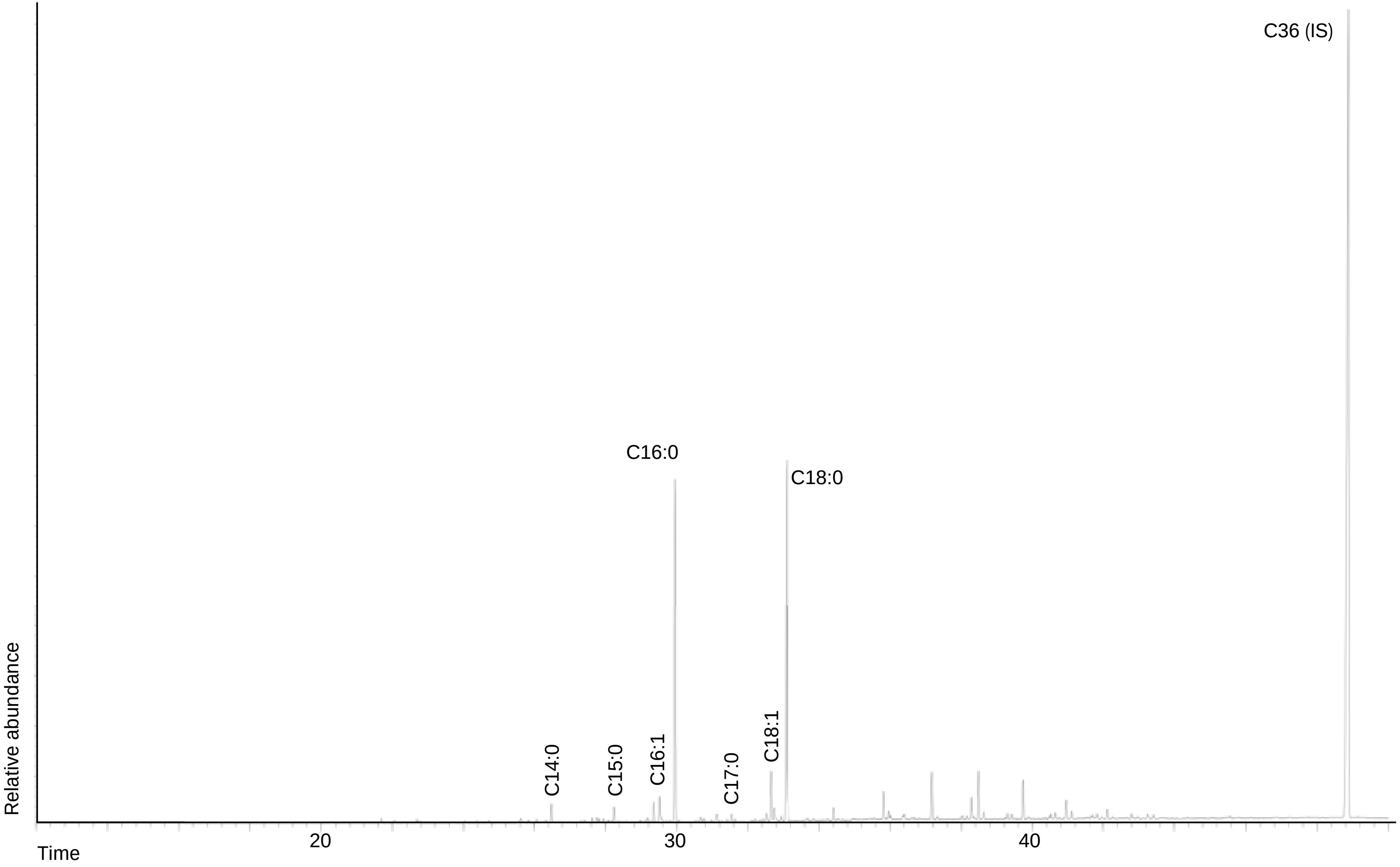
GVP1


C36 (IS)
C18:0
C16:0
C18:1
Phytanic acid
C22:1
C14:0
C12:0
C15:0
C17:0br
C17:0
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
C25:0
C26:0
Relative abundance
Time
20
30
40

GVP2

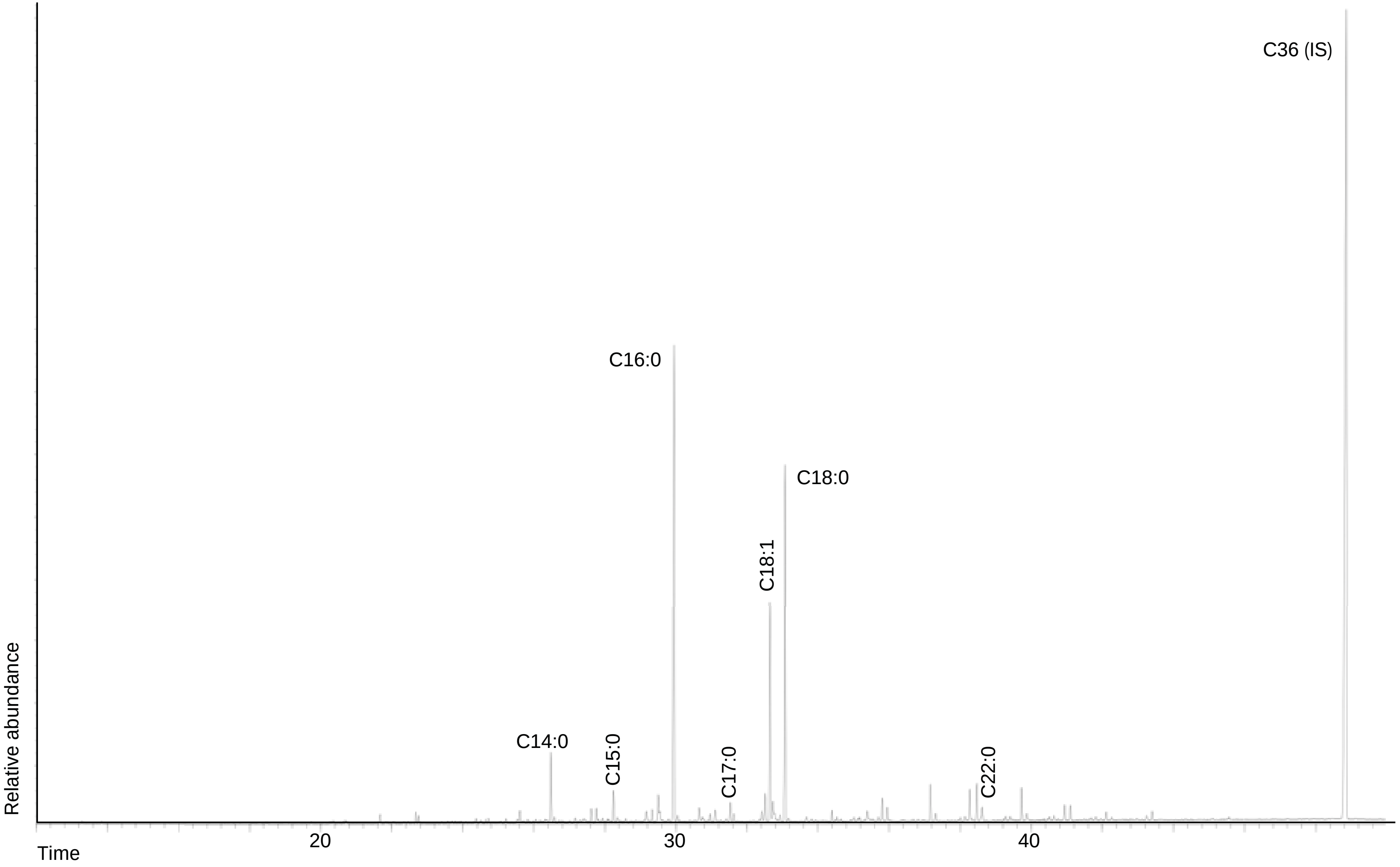

C36 (IS)
C18:0
C16:0
D9
D10
C12:0
C13:0
C14:0
C15:0
C18:1
C17:br
C17:0
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
C24:0
Relative abundance
Time
20
30
40

GVP3

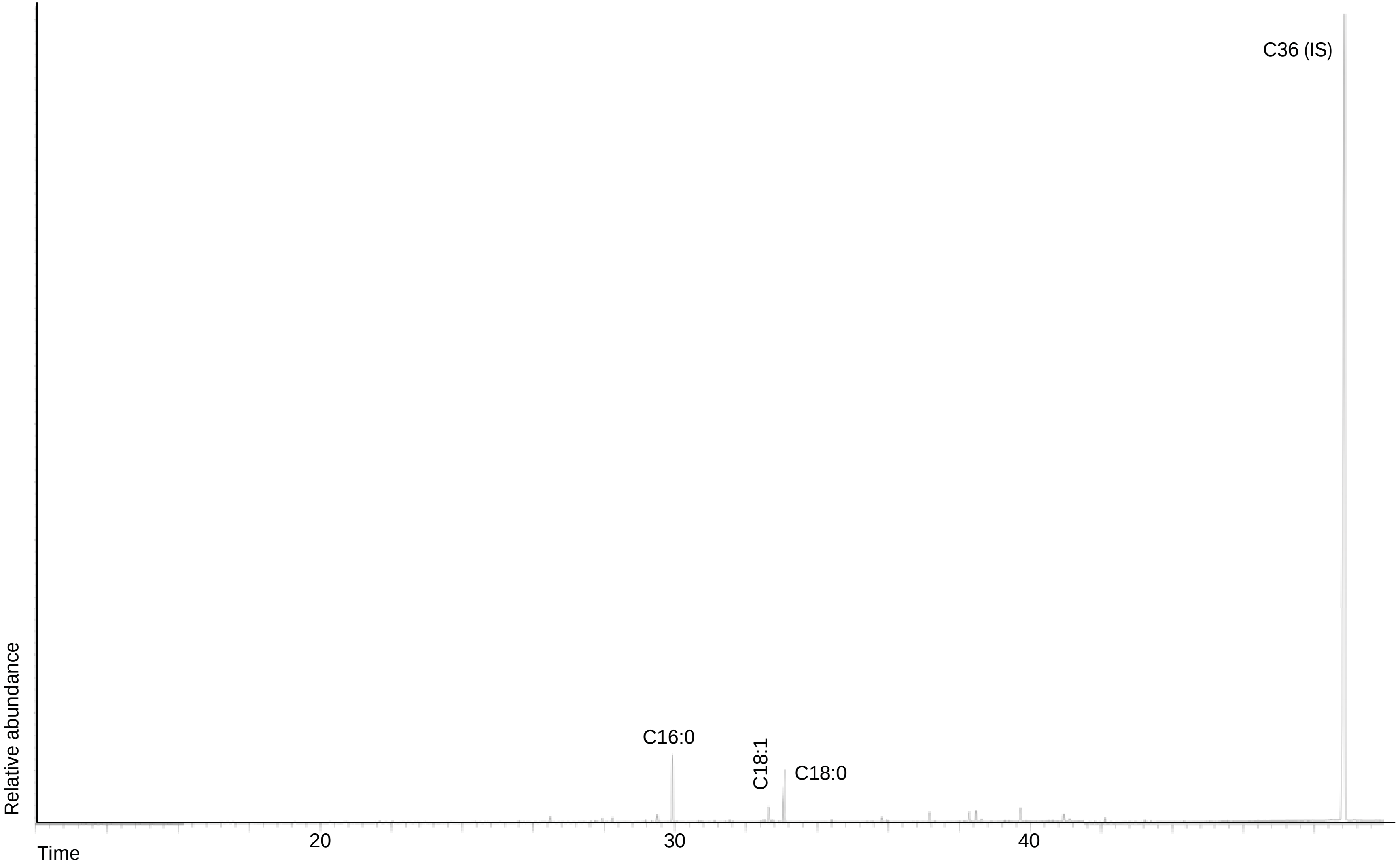


C36 (IS)
C16:0
C18:0
C14:0
C15:0
C16:1
C17:0
C18:1
Relative abundance
Time
20
30
40

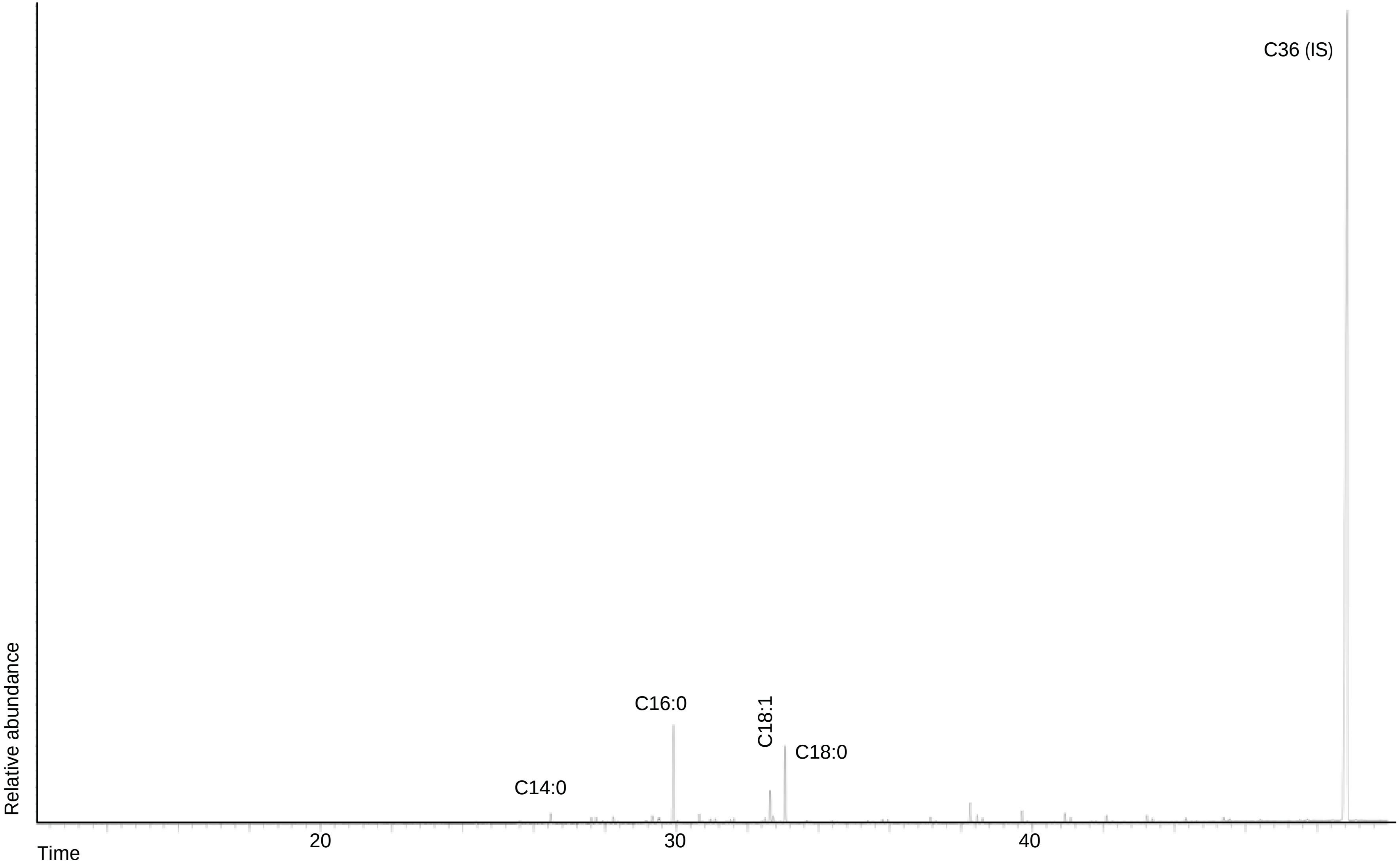
GVP4


C36 (IS)
C16:0
C18:0
C18:1
C14:0
C15:0
C17:0
C22:0
Relative abundance
Time
20
30
40

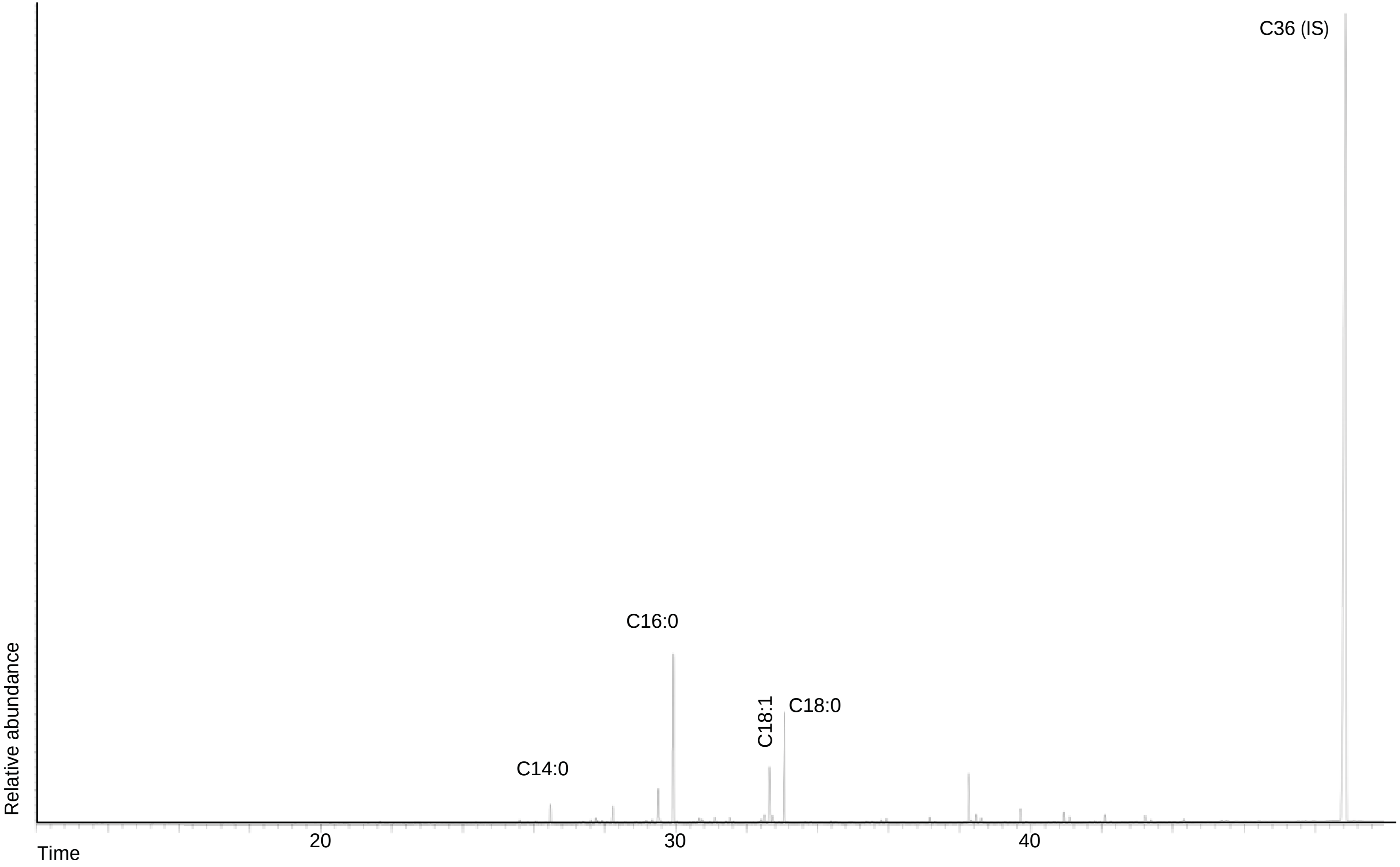
GVP5



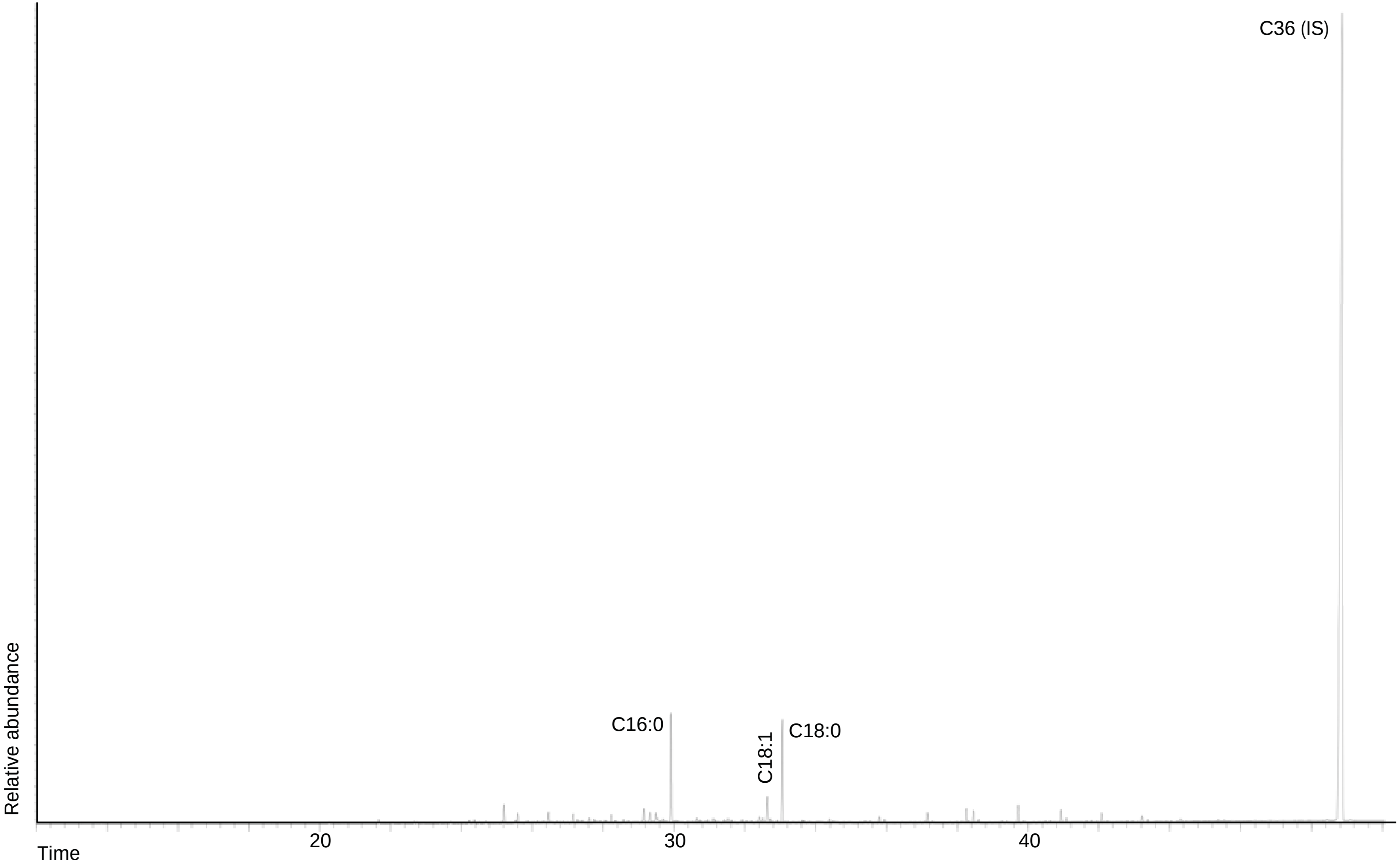
GV101



GV28I

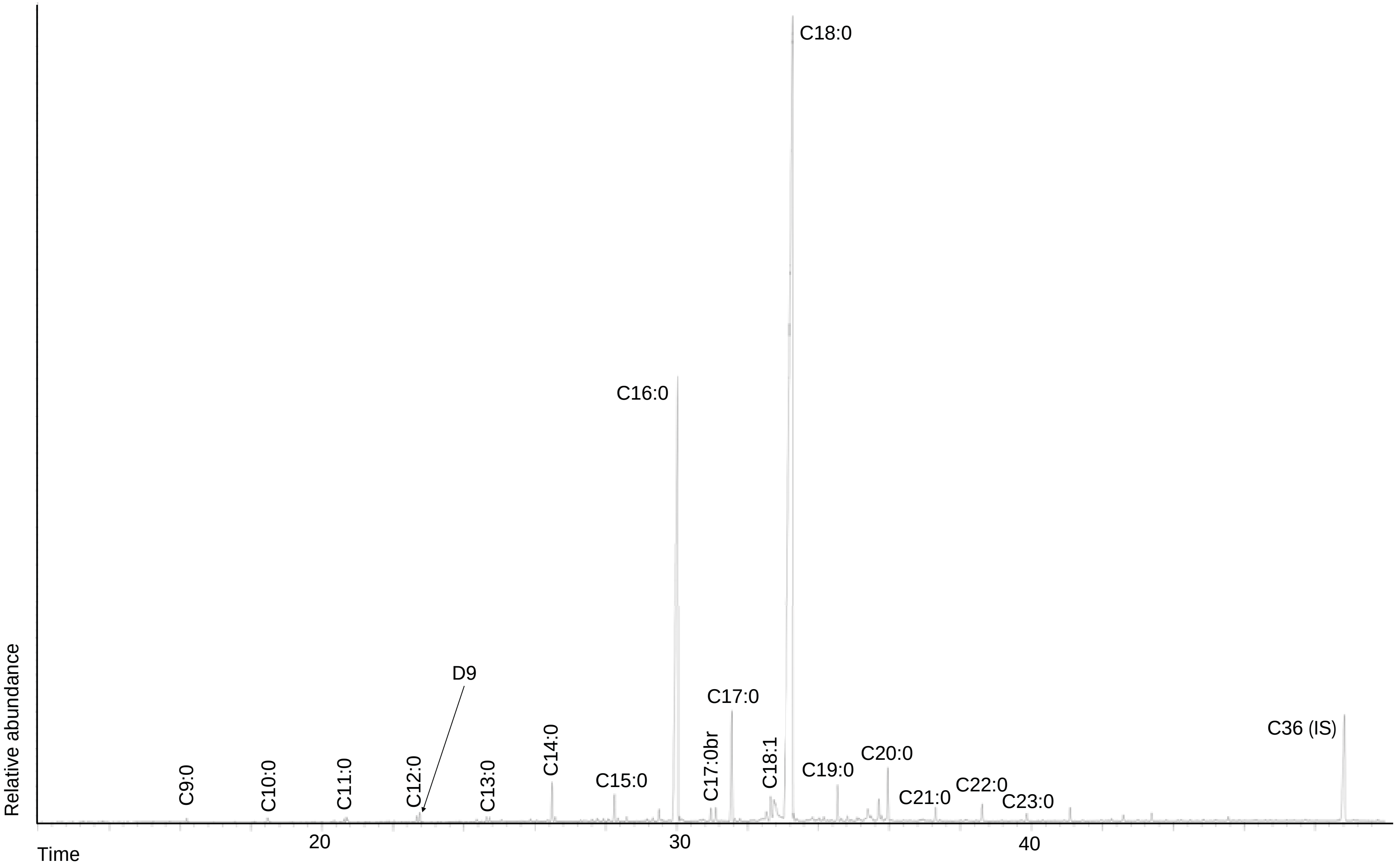


GV54

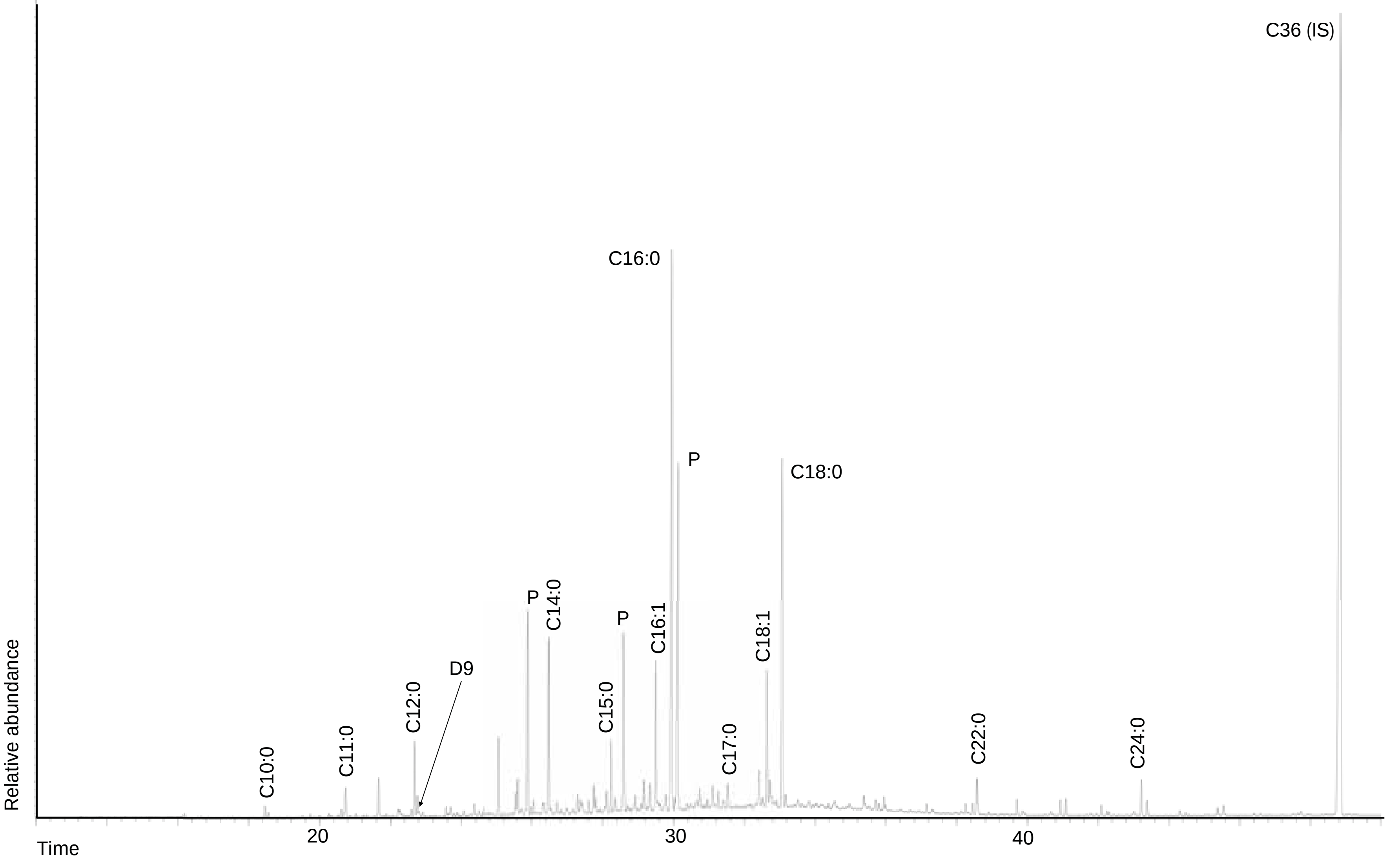


Cova de la Guineu

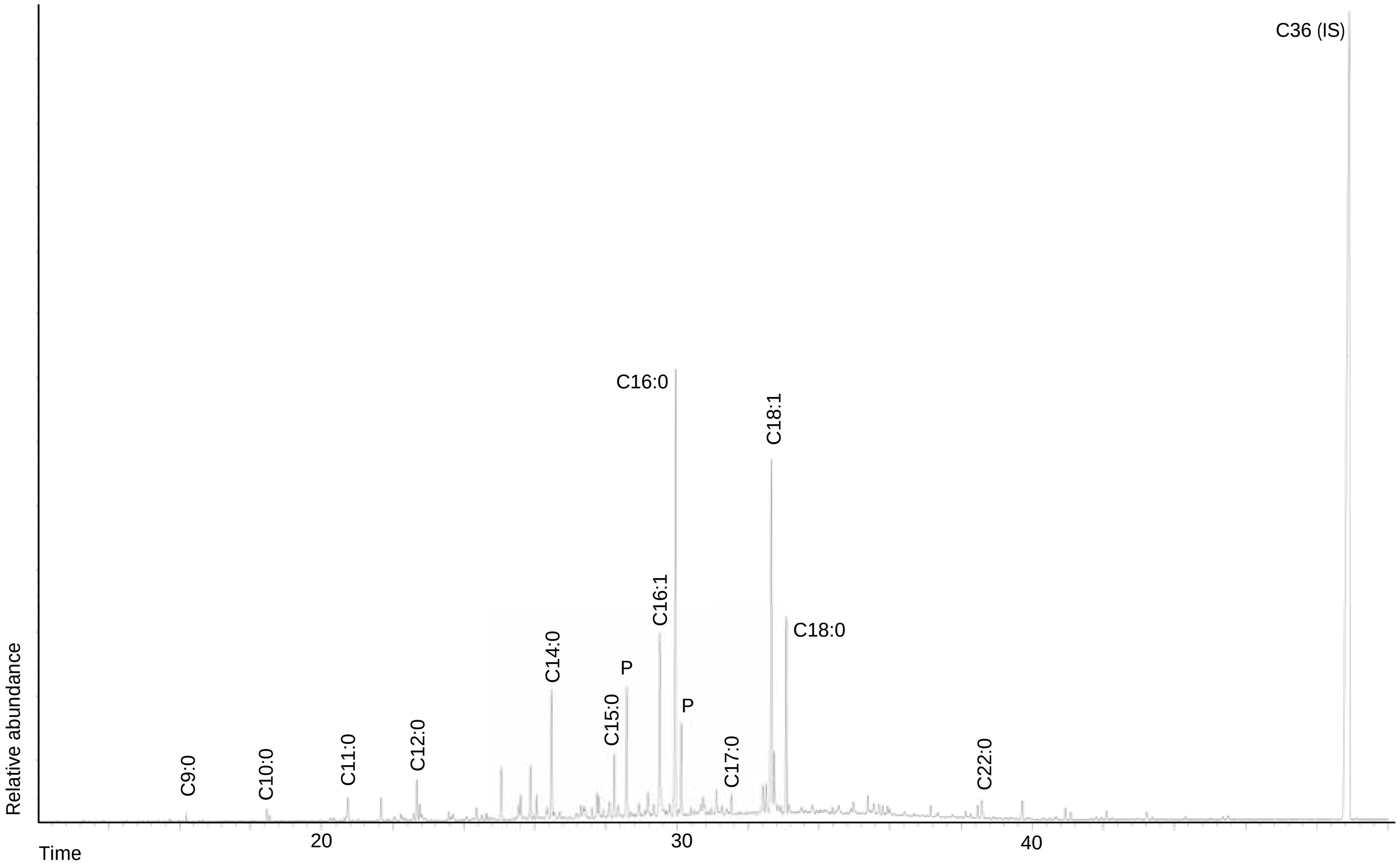
CG1


C18:0
C16:0
D9
C17:0
C36 (IS)
C17:0br
C18:1
C20:0
C14:0
C19:0
C15:0
C22:0
C21:0
C23:0
C9:0
C10:0
C11:0
C12:0
C13:0
Relative abundance
Time
20
30
40

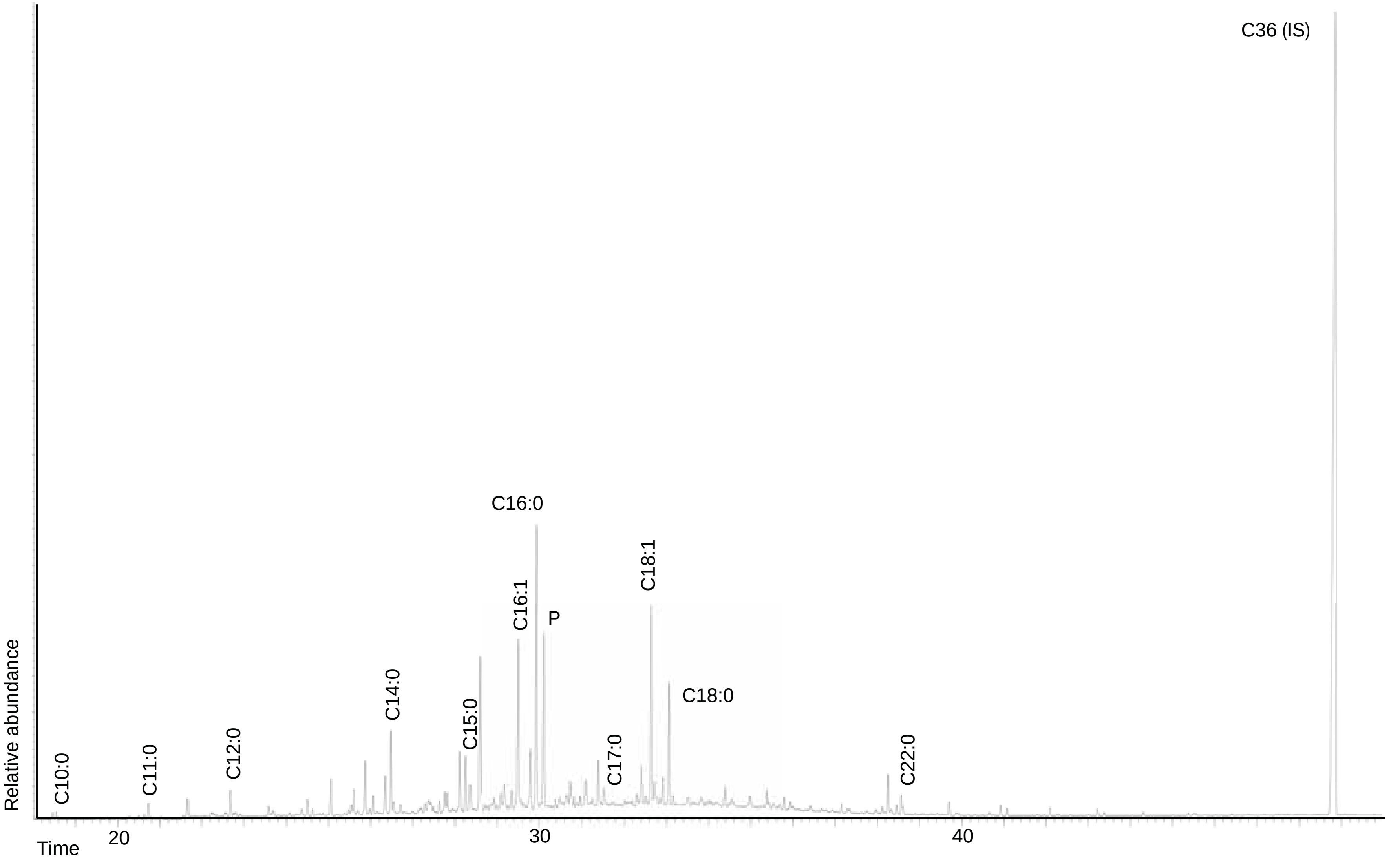
CG2


C36 (IS)
C16:0
P
C18:0
P
C14:0
P
C16:1
C18:1
D9
C12:0
C15:0
C11:0
C10:0
C17:0
C22:0
C24:0
Relative abundance
Time
20
30
40

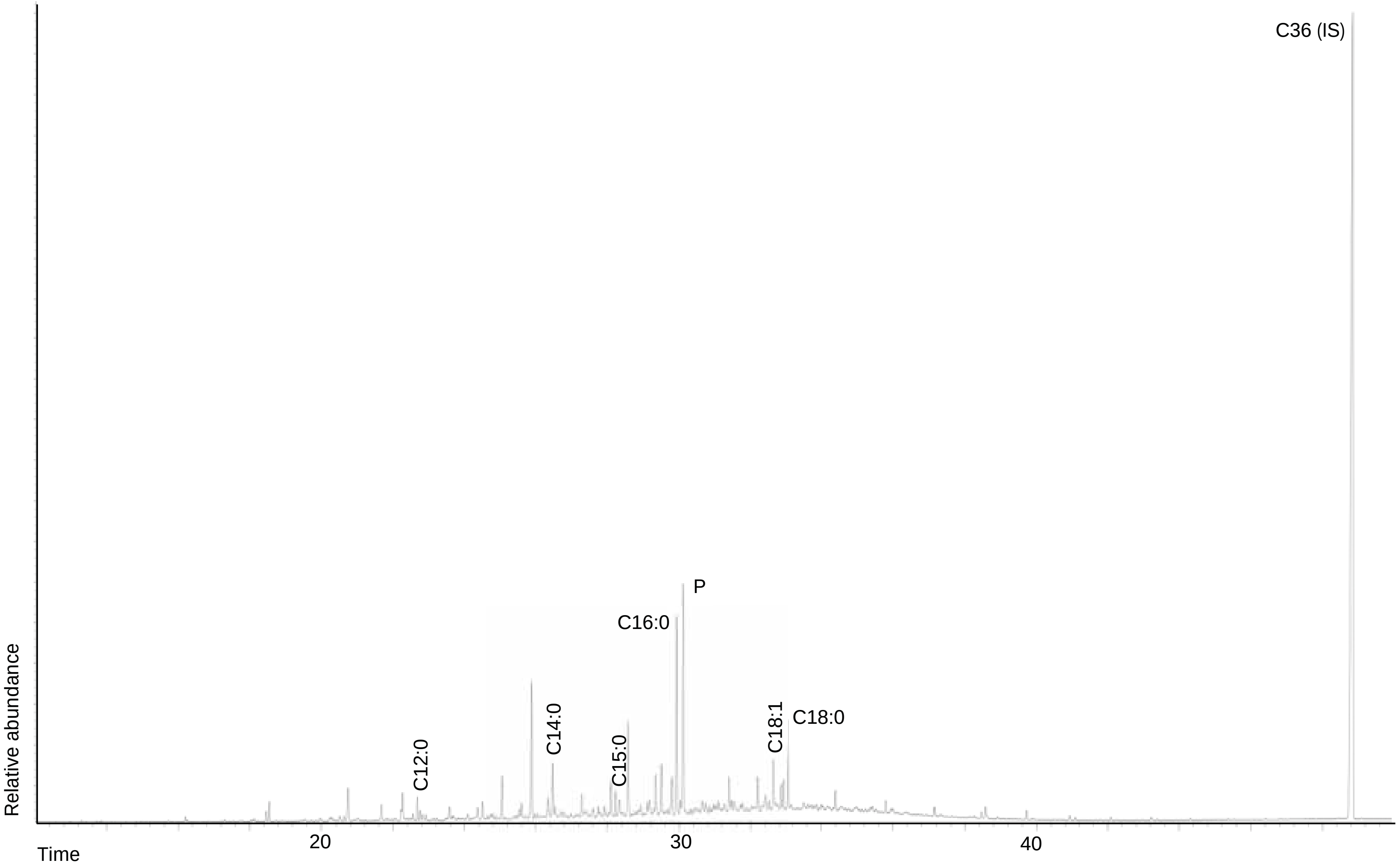
CG3


C36 (IS)
C16:0
C18:1
C16:1
C18:0
C14:0
P
P
C15:0
C12:0
C11:0
C17:0
C22:0
C10:0
C9:0
Relative abundance
Time
20
30
40

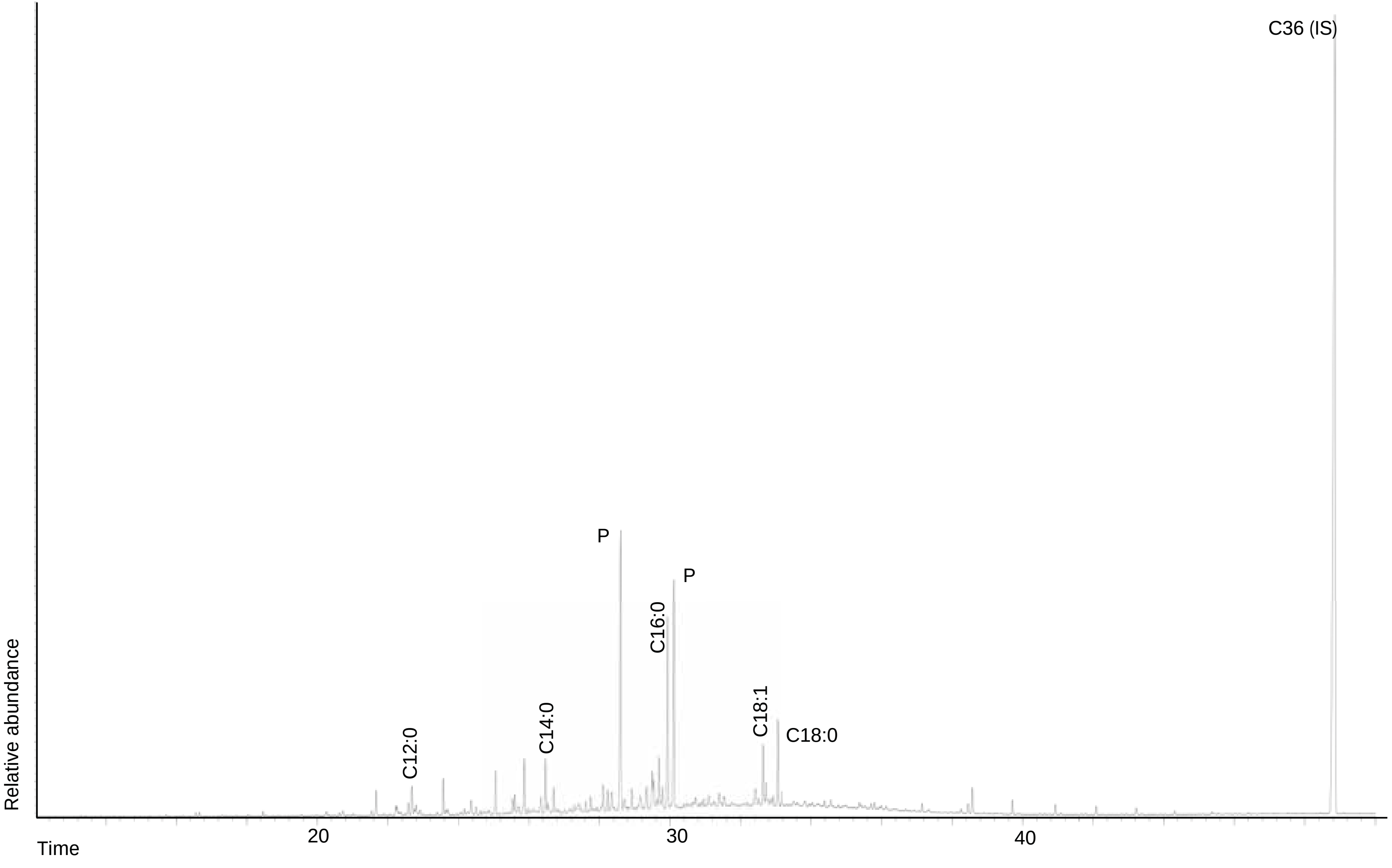
CG17


C36 (IS)
C16:0
C18:1
C16:1
P
C18:0
C14:0
C15:0
C17:0
C22:0
C12:0
C11:0
C10:0
Relative abundance
Time
20
30
40

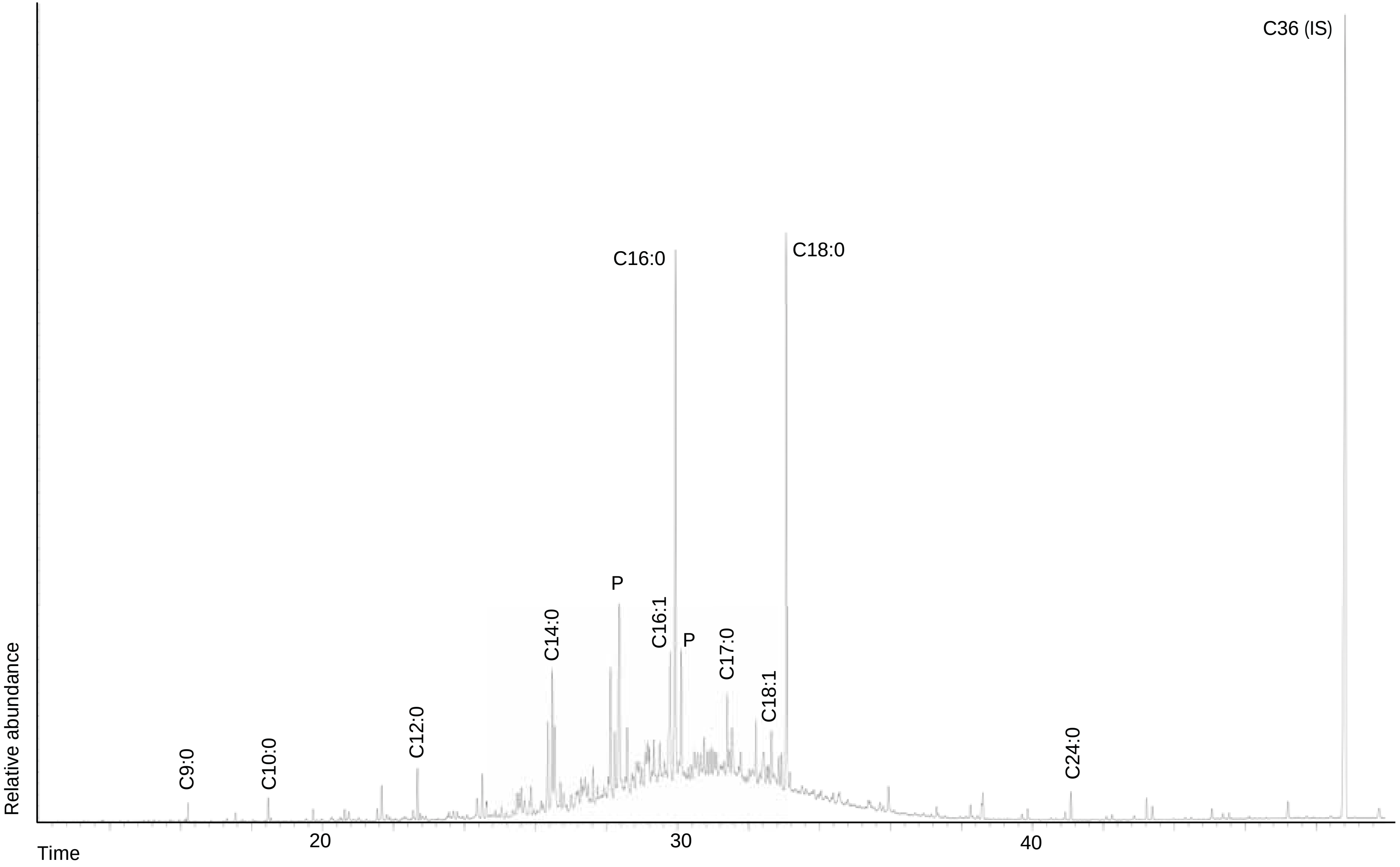
CG19


C36 (IS)
P
C16:0
C12:0
C14:0
C15:0
C18:1
C18:0
Relative abundance
Time
20
30
40

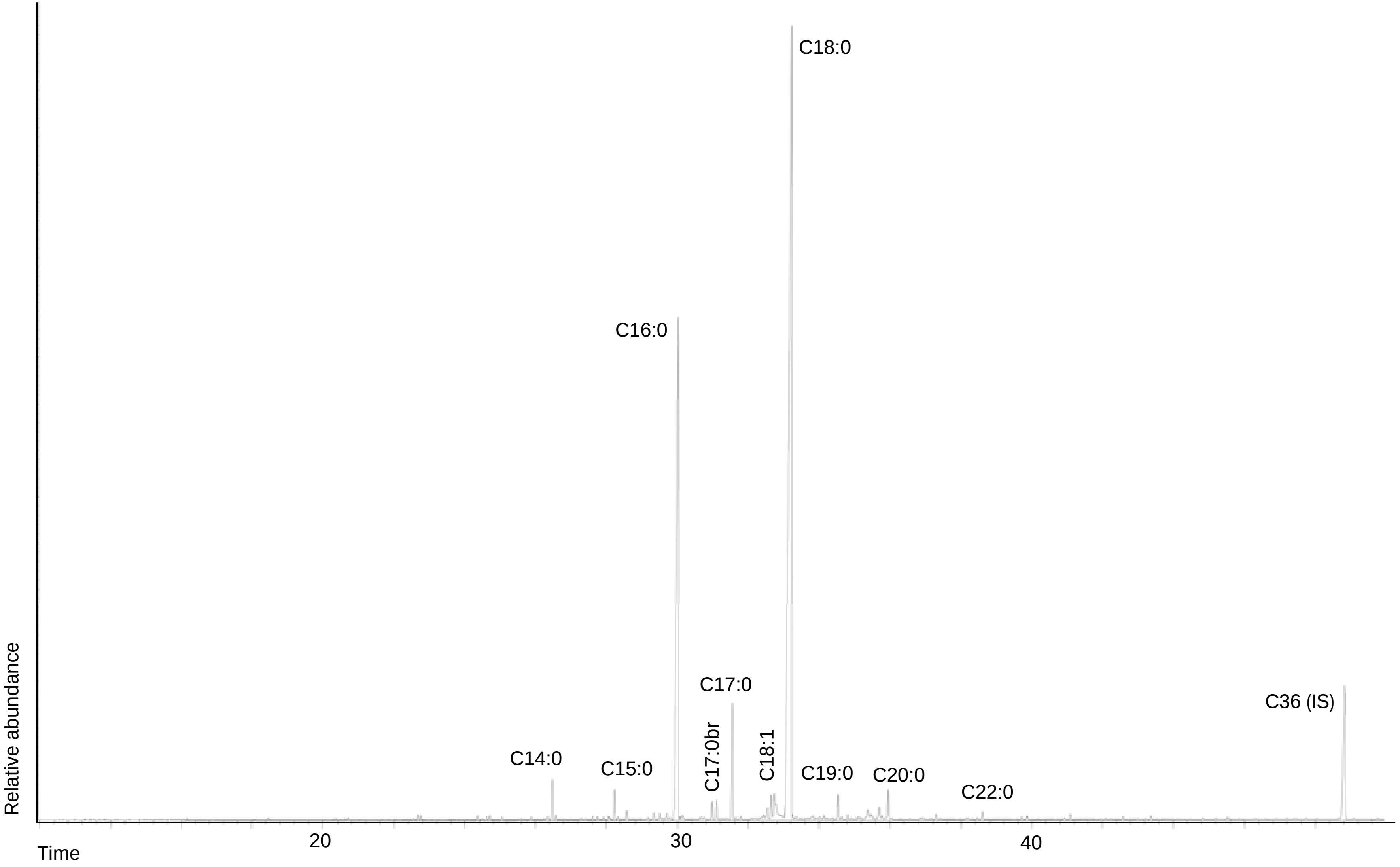
CG20


C36 (IS)
P
P
C16:0
C12:0
C14:0
C18:1
C18:0
Relative abundance
Time
20
30
40

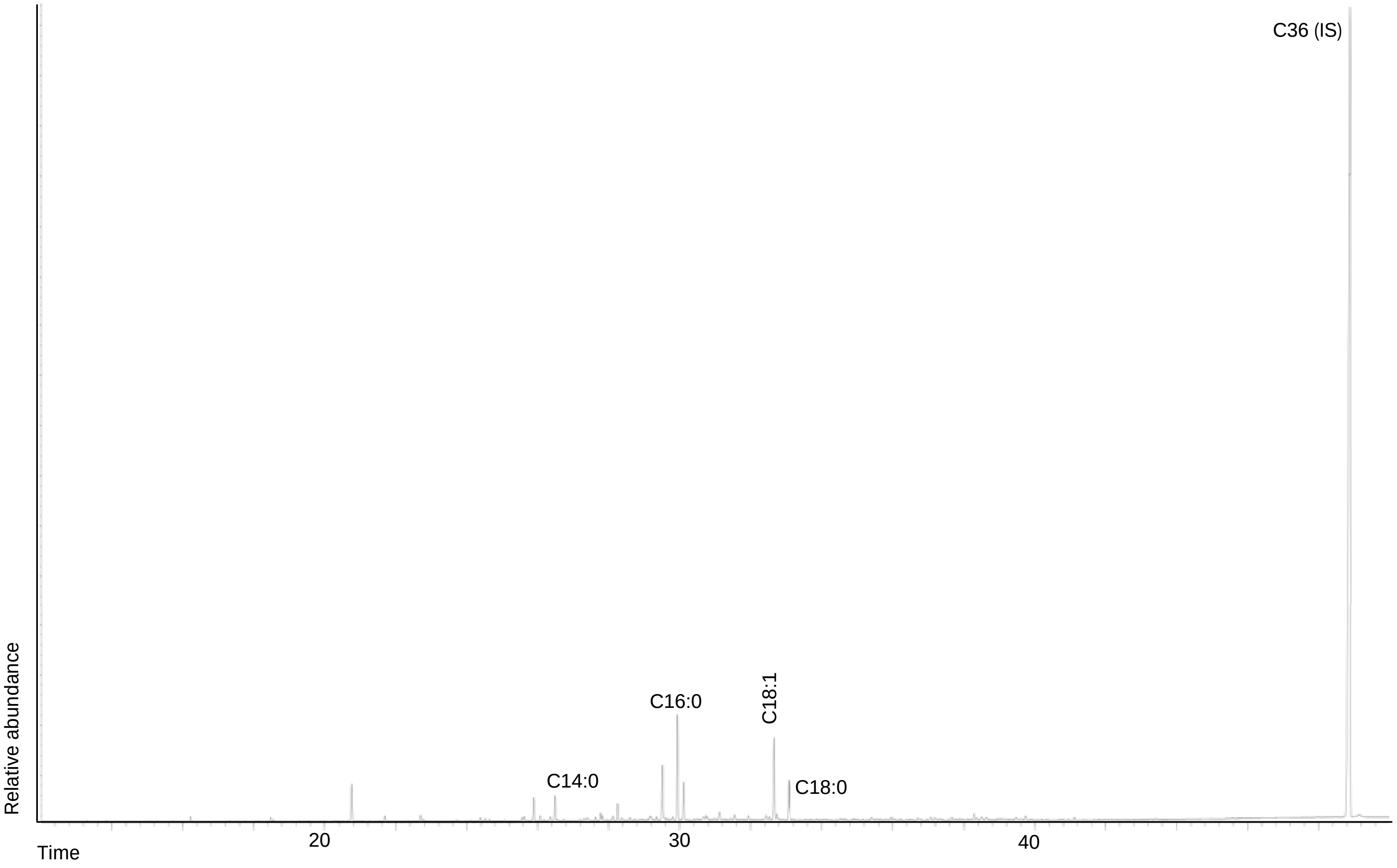
CG26


C36 (IS)
C16:0
C18:0
P
C14:0
C16:1
P
C17:0
C18:1
C12:0
C9:0
C10:0
C24:0
Relative abundance
Time
20
30
40

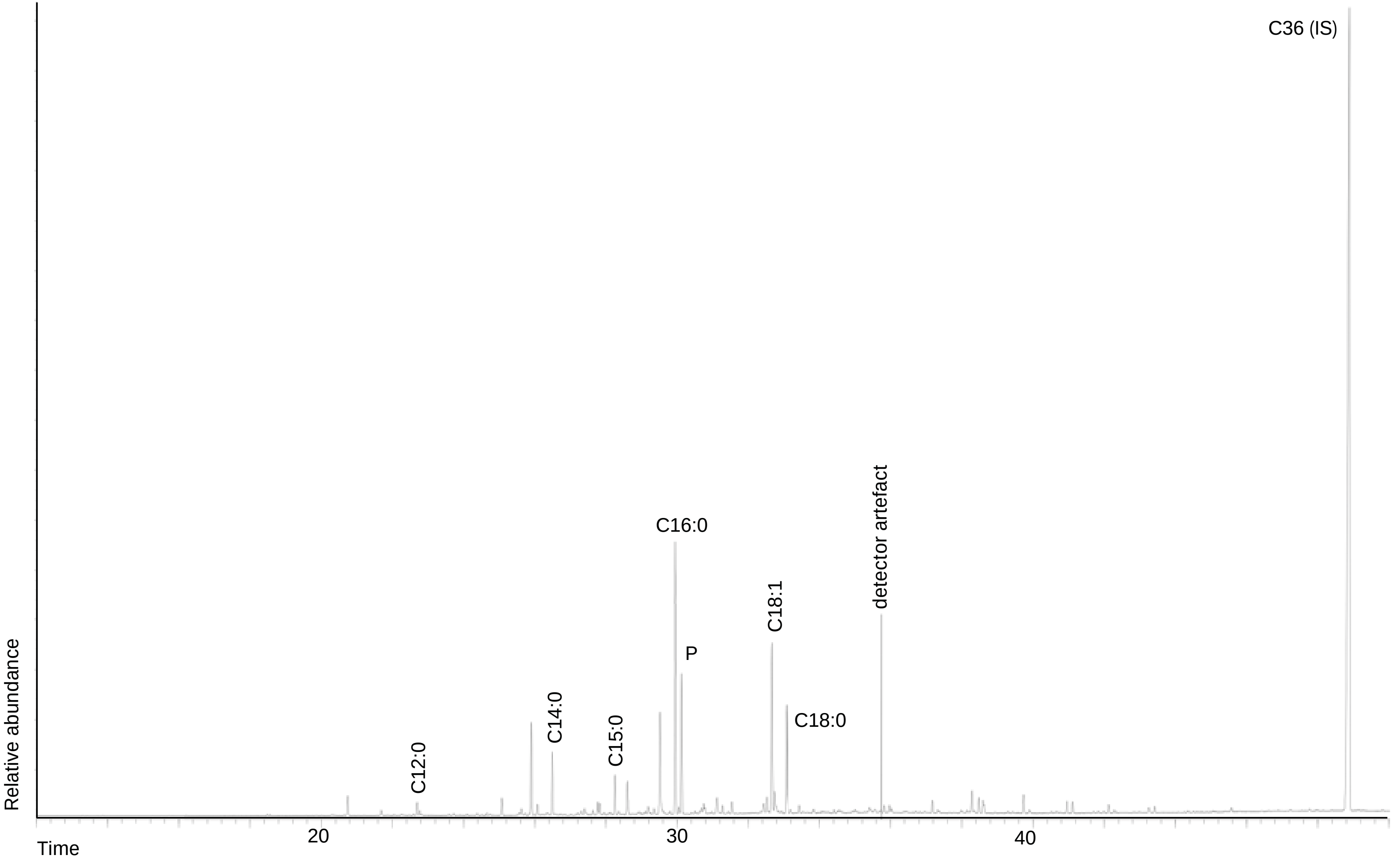
CG27


C18:0
C16:0
C17:0
C36 (IS)
C17:0br
C18:1
C14:0
C15:0
C19:0
C20:0
C22:0
Relative abundance
Time
20
30
40

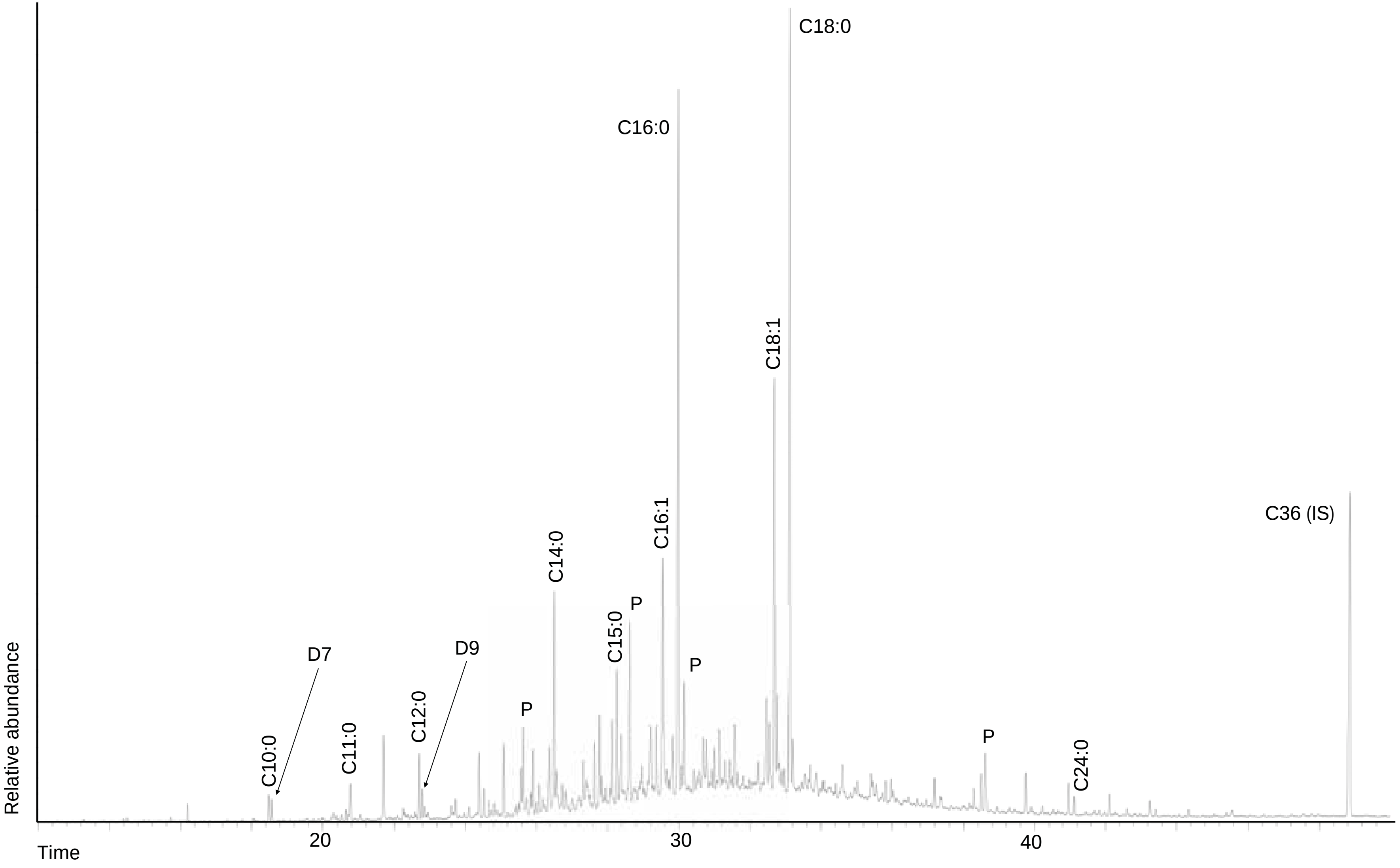
CGP1



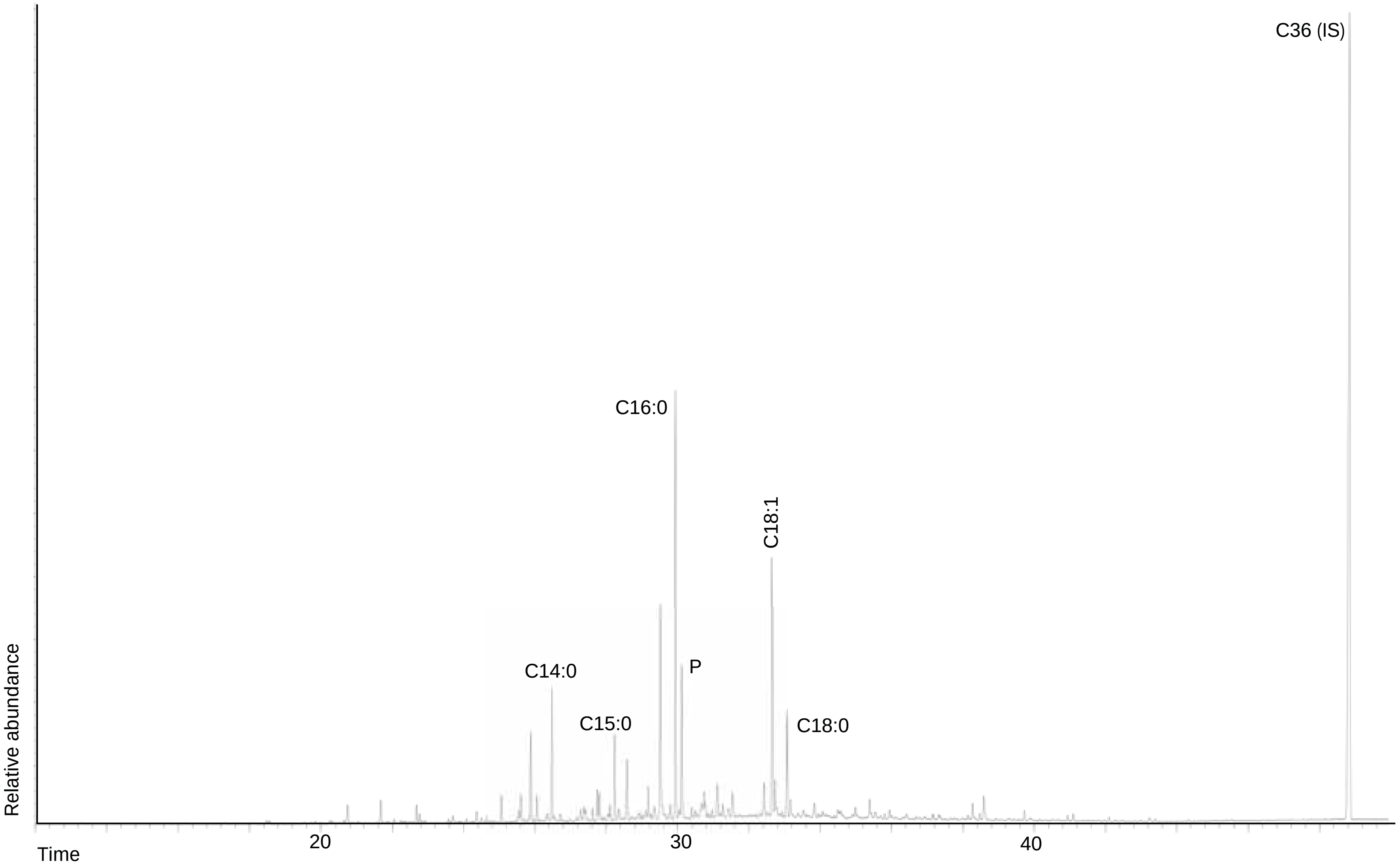
CGP2


C36 (IS)
C16:0
detector artefact
C18:1
P
C18:0
C14:0
C15:0
C12:0
Relative abundance
Time
20
30
40

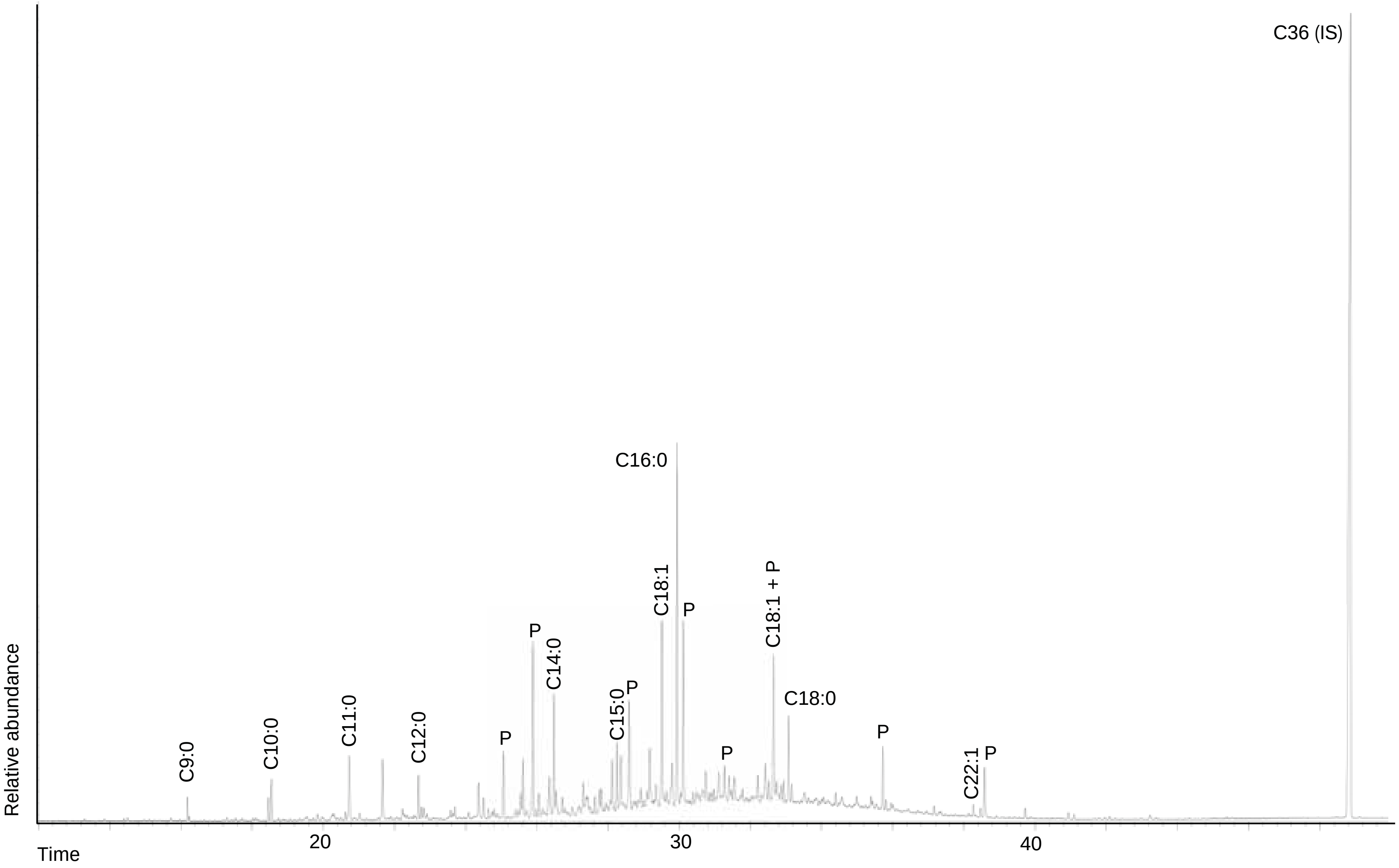
CGP3


C18:0
C16:0
C18:1
C36 (IS)
C16:1
C14:0
P
C15:0
D7
D9
P
P
C12:0
C11:0
C10:0
P
C24:0
Relative abundance
Time
20
30
40

CGP4


C36 (IS)
C16:0
C18:1
P
C14:0
C15:0
C18:0
Relative abundance
Time
20
30
40

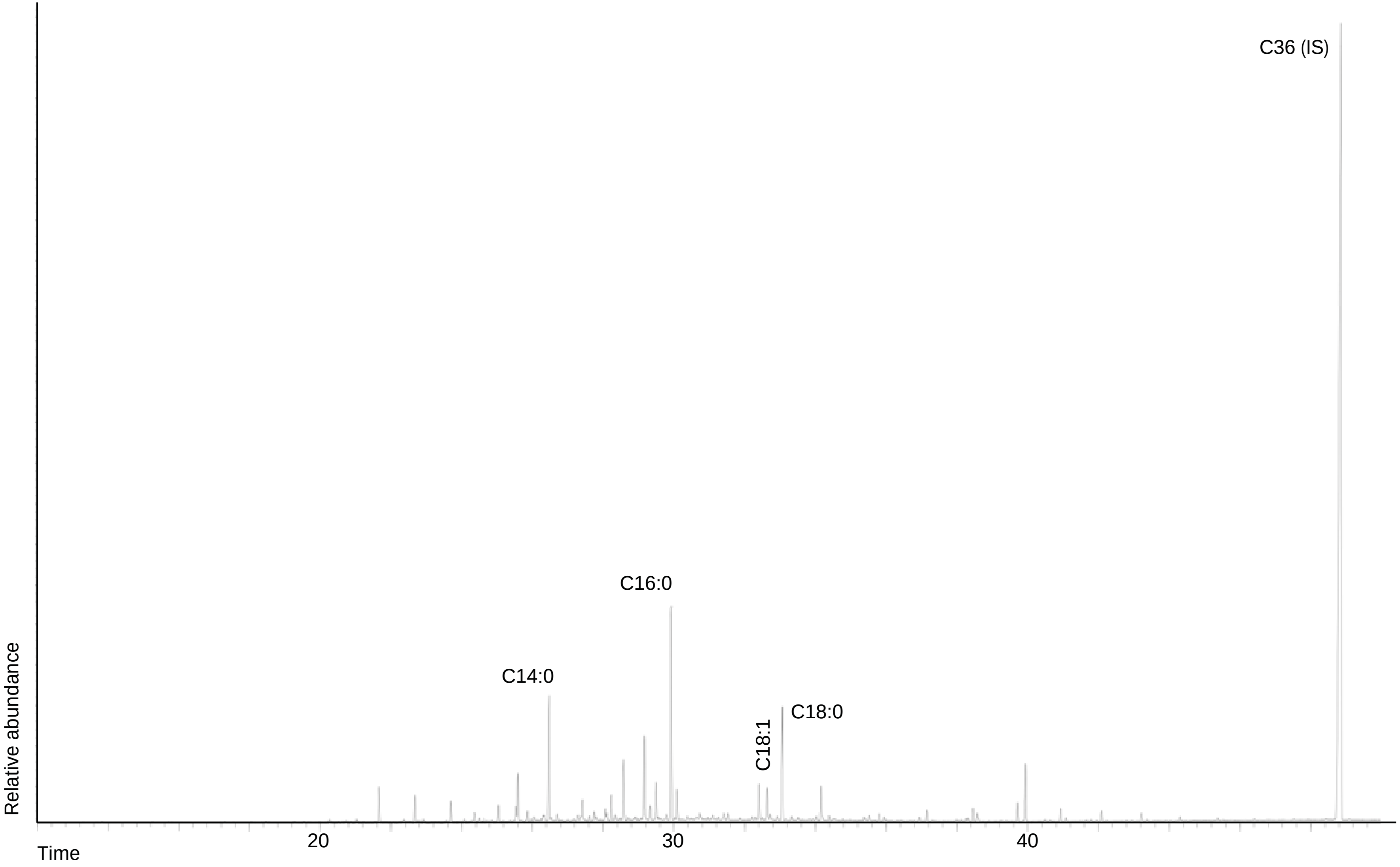
CGP5



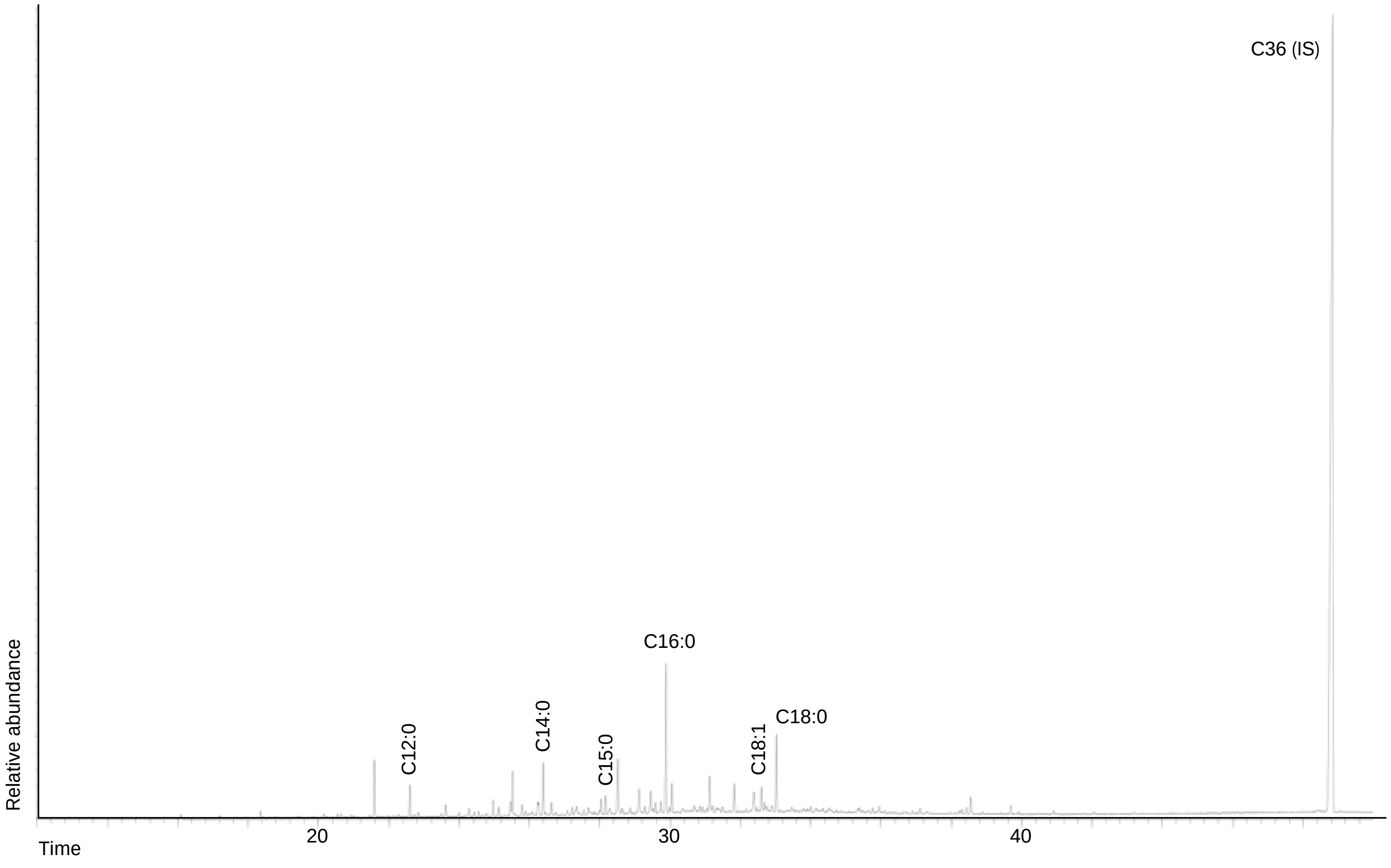
C36 (IS)
C16:0
C18:1
P
C18:1 + P
P
C14:0
C15:0
P
C18:0
P
C9:0
C10:0
C11:0
C12:0
P
P
C22:1
P
Relative abundance
Time
20
30
40

El Cavet

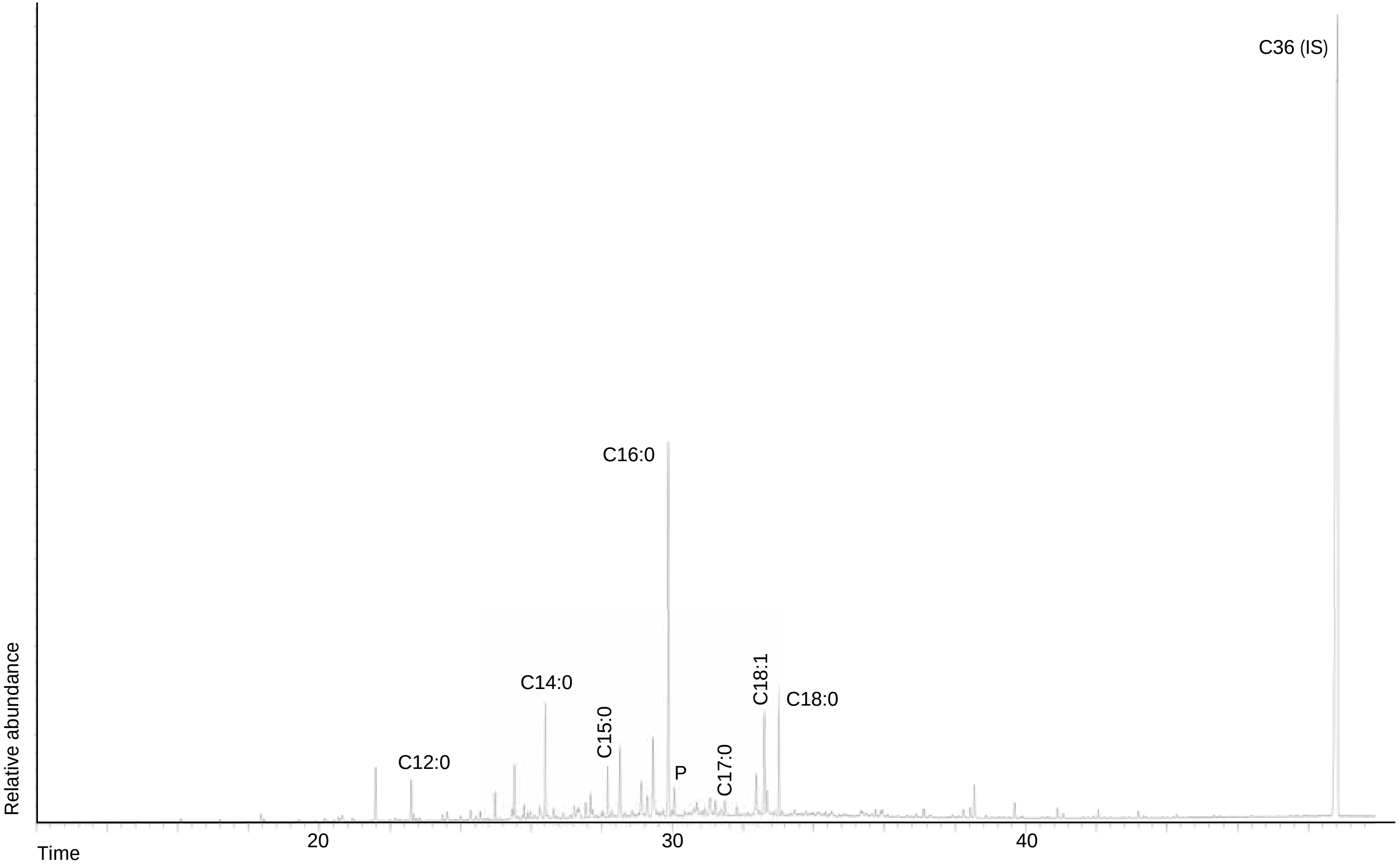
CV25



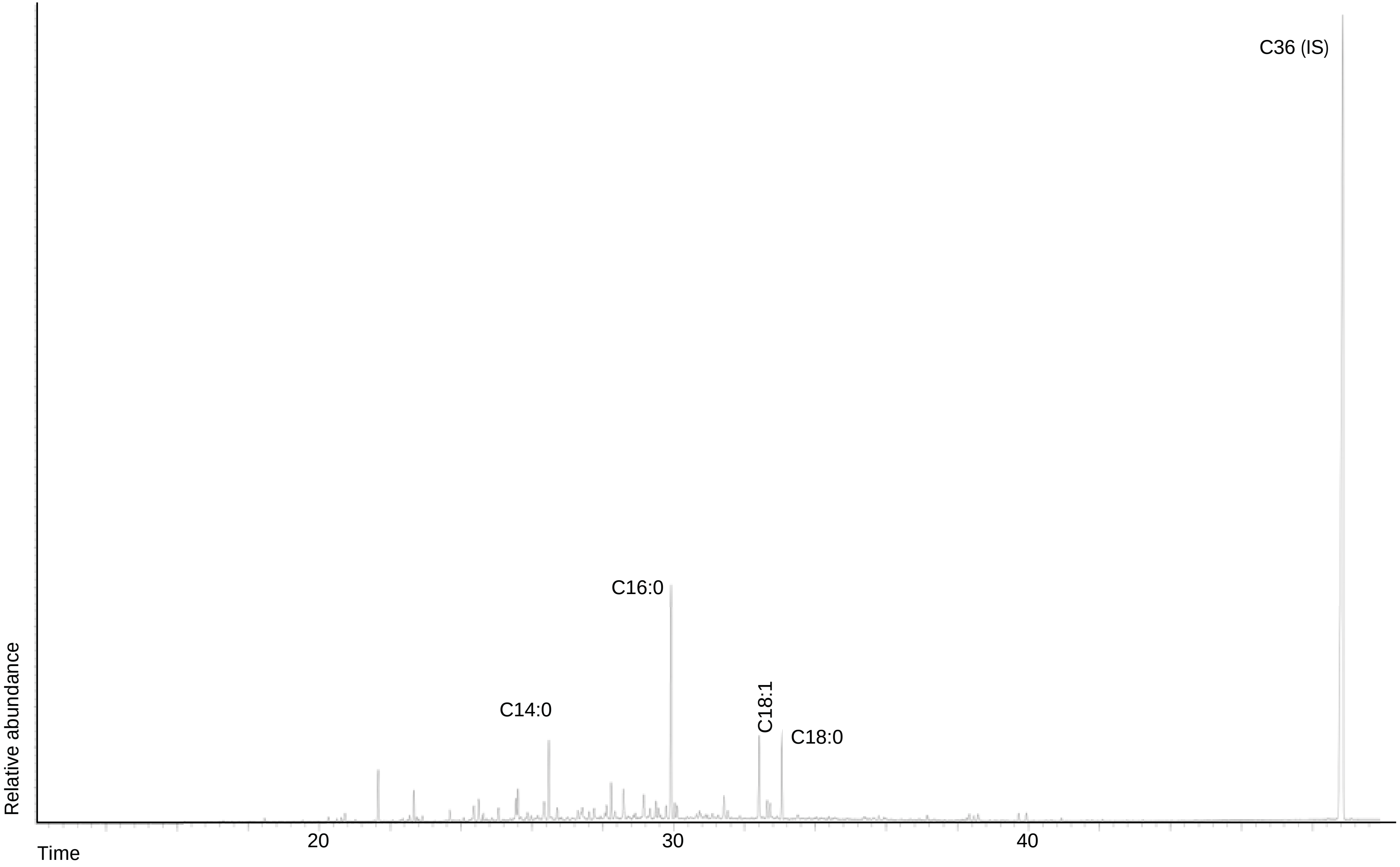
CV36


C36 (IS)
C16:0
C18:0
C12:0
C14:0
C15:0
C18:1
Relative abundance
Time
20
30
40

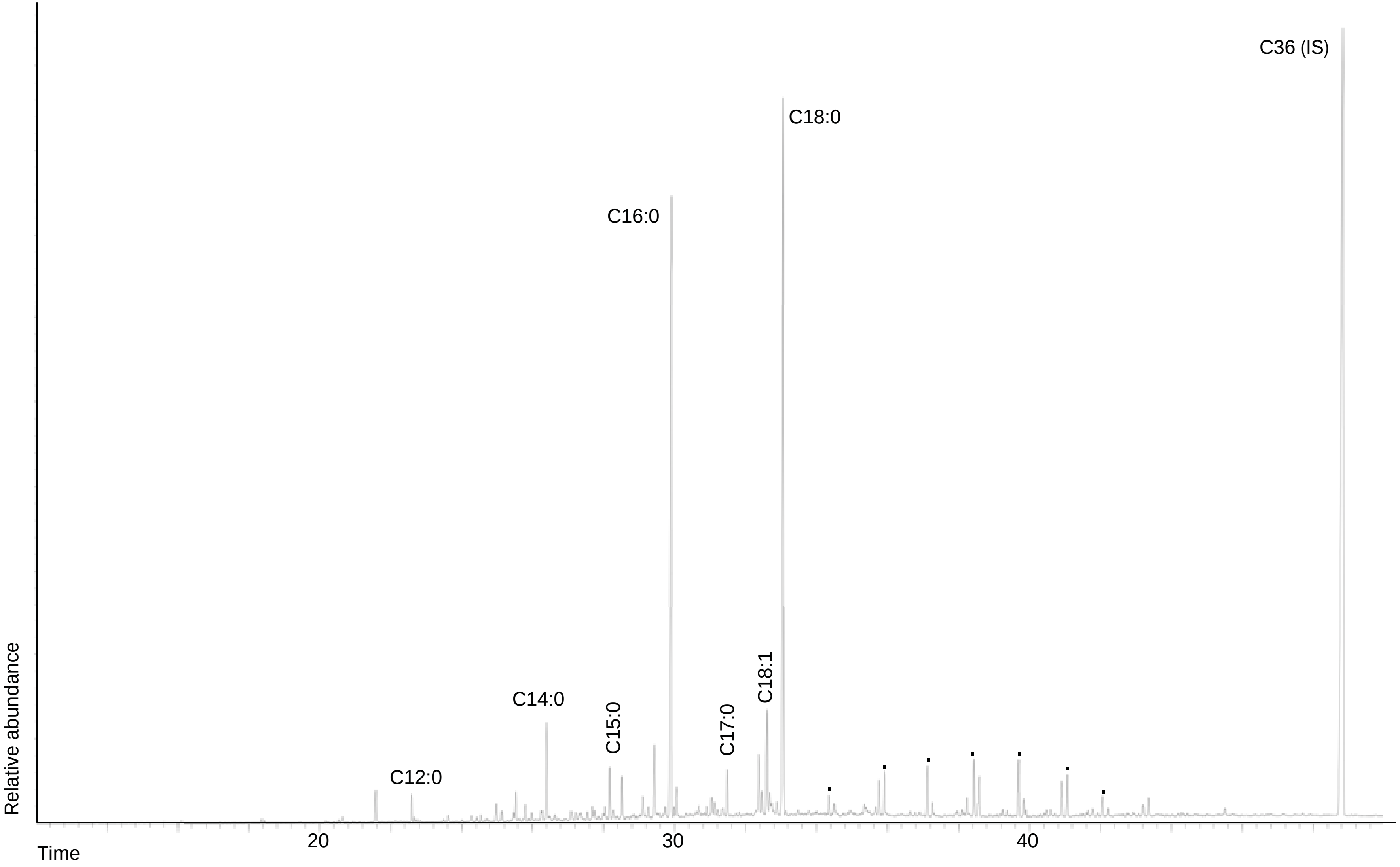
CV37


Relative abundance
Time
20
30
40
C12:0
C14:0
C15:0
C16:0
P
C17:0
C18:1
C18:0
C36 (IS)

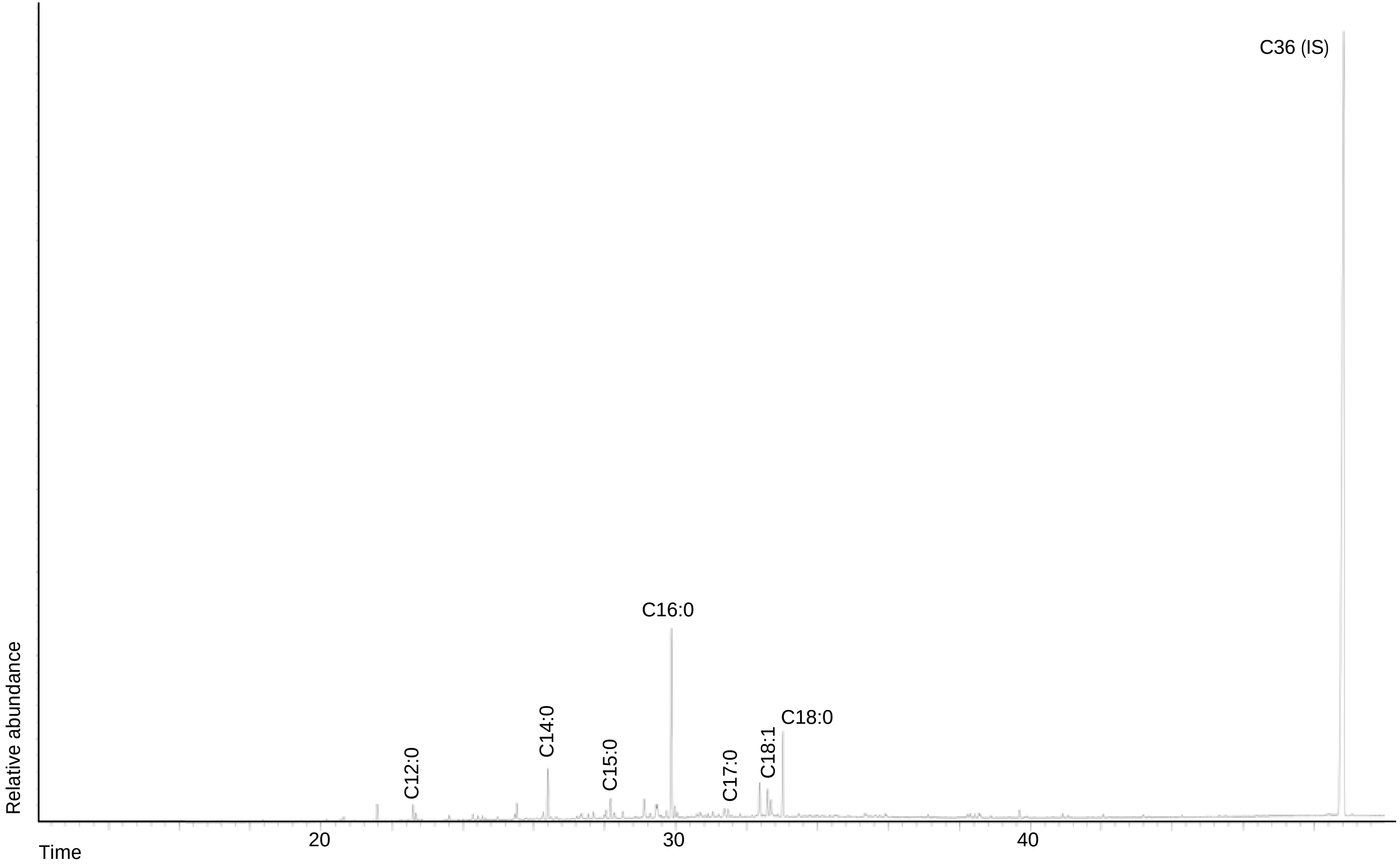
CV56



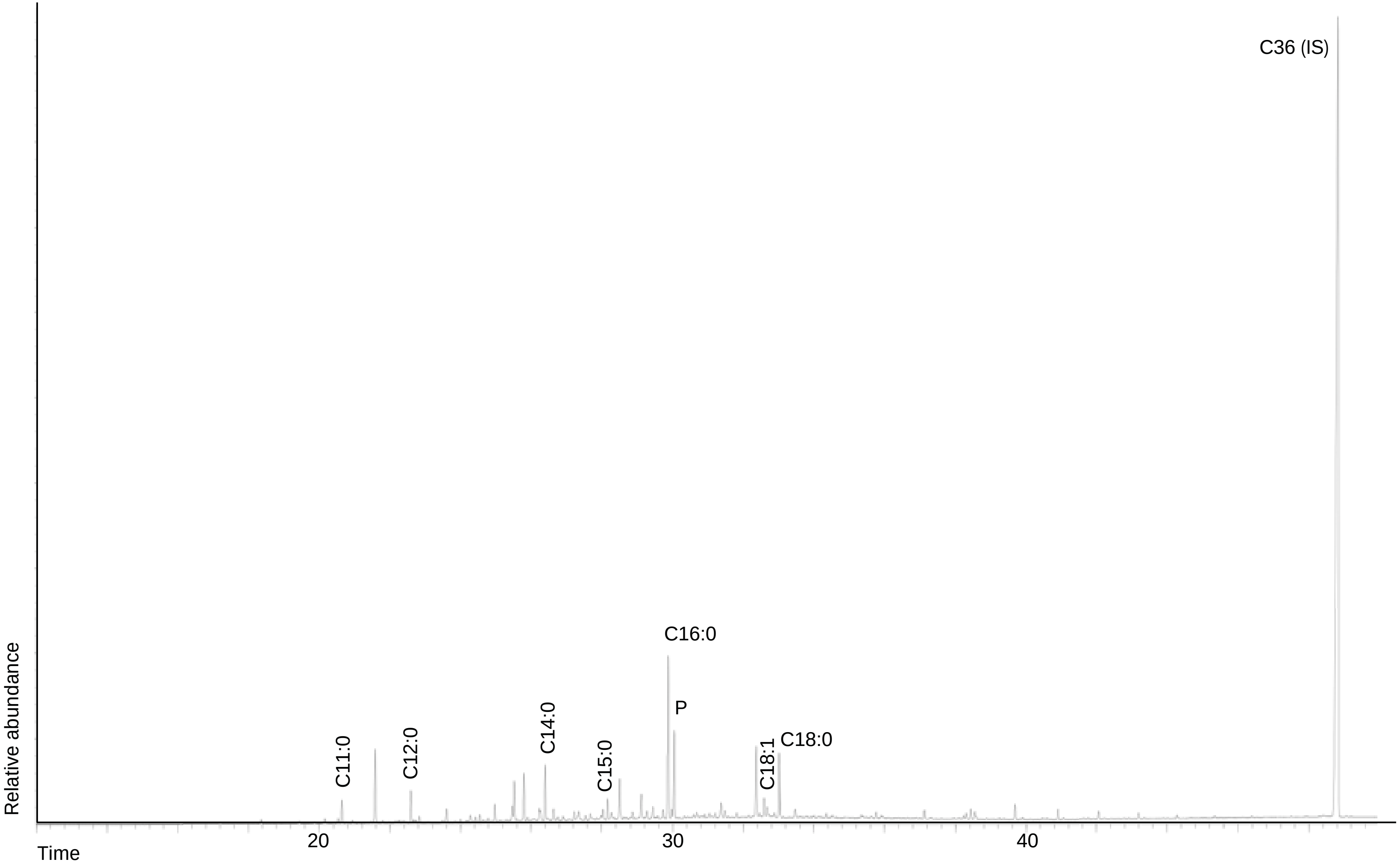
CV60


C36 (IS)
C18:0
C16:0
C18:1
C14:0
C15:0
C17:0
C12:0
Relative abundance
Time
20
30
40

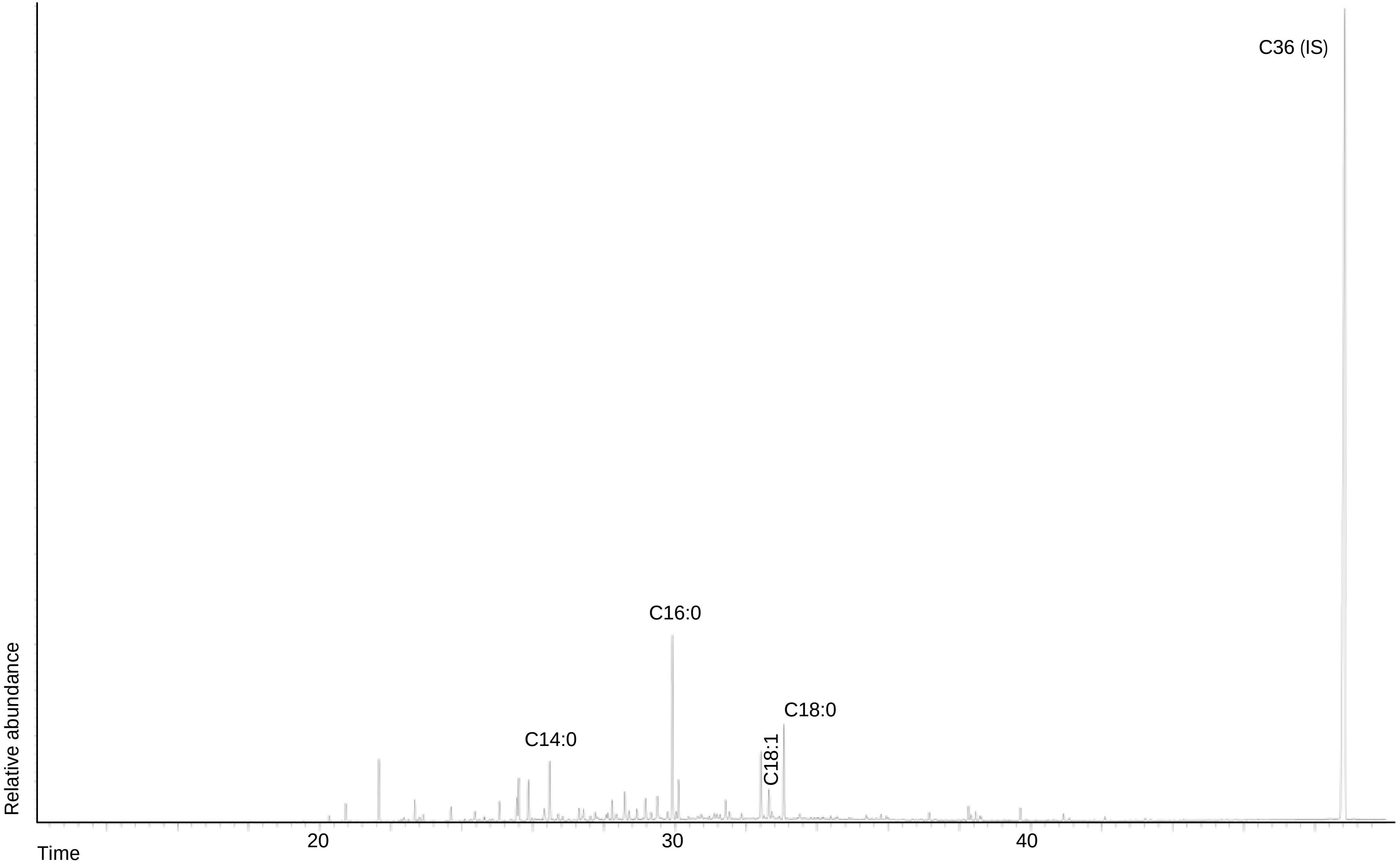
CV66


C36 (IS)
C16:0
C18:0
C14:0
C12:0
C15:0
C17:0
C18:1
Relative abundance
20
30
40
Time

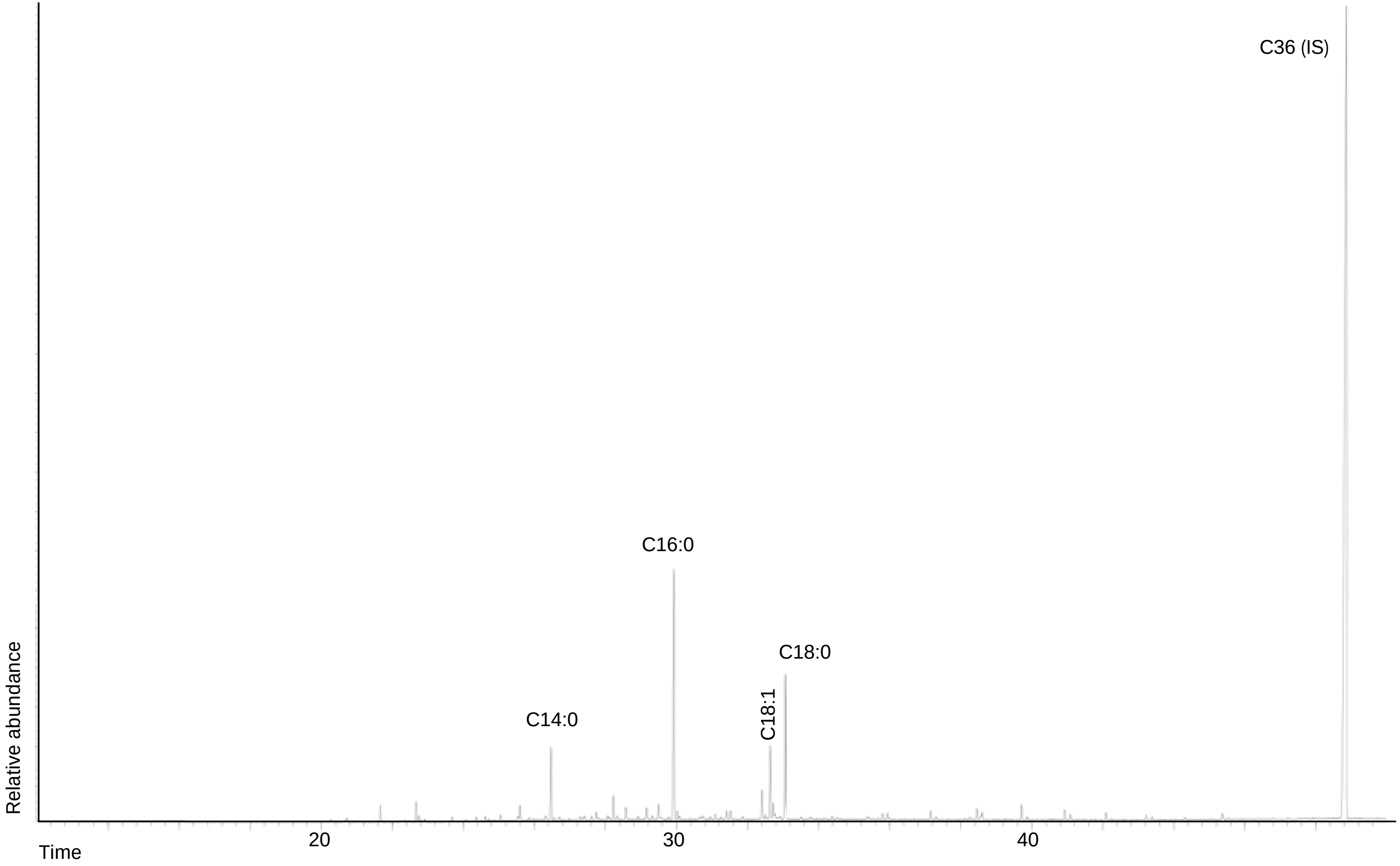
CV114


C36 (IS)
C16:0
P
C11:0
C12:0
C14:0
C15:0
C18:1
C18:0
Relative abundance
20
30
40
Time

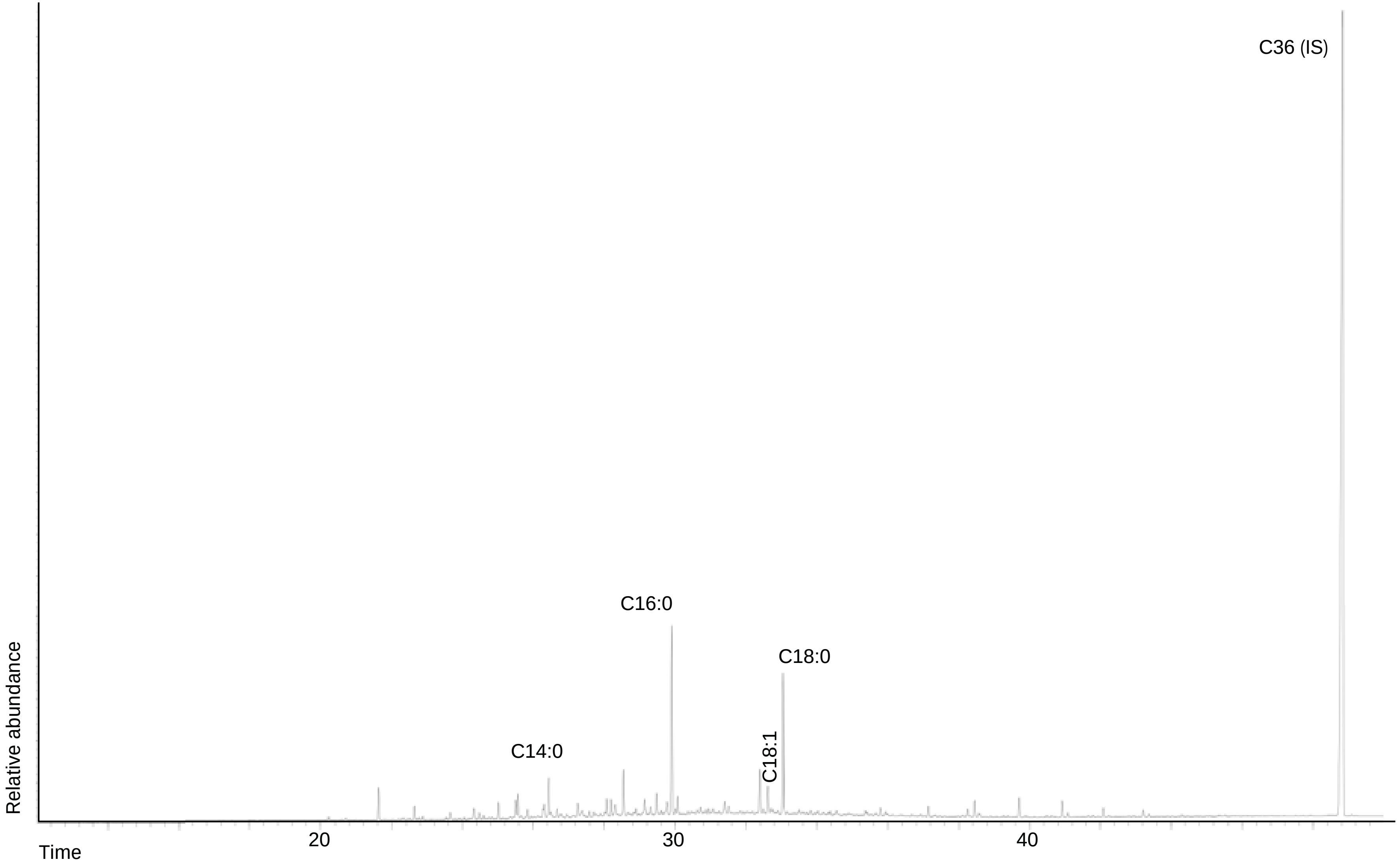
CV164



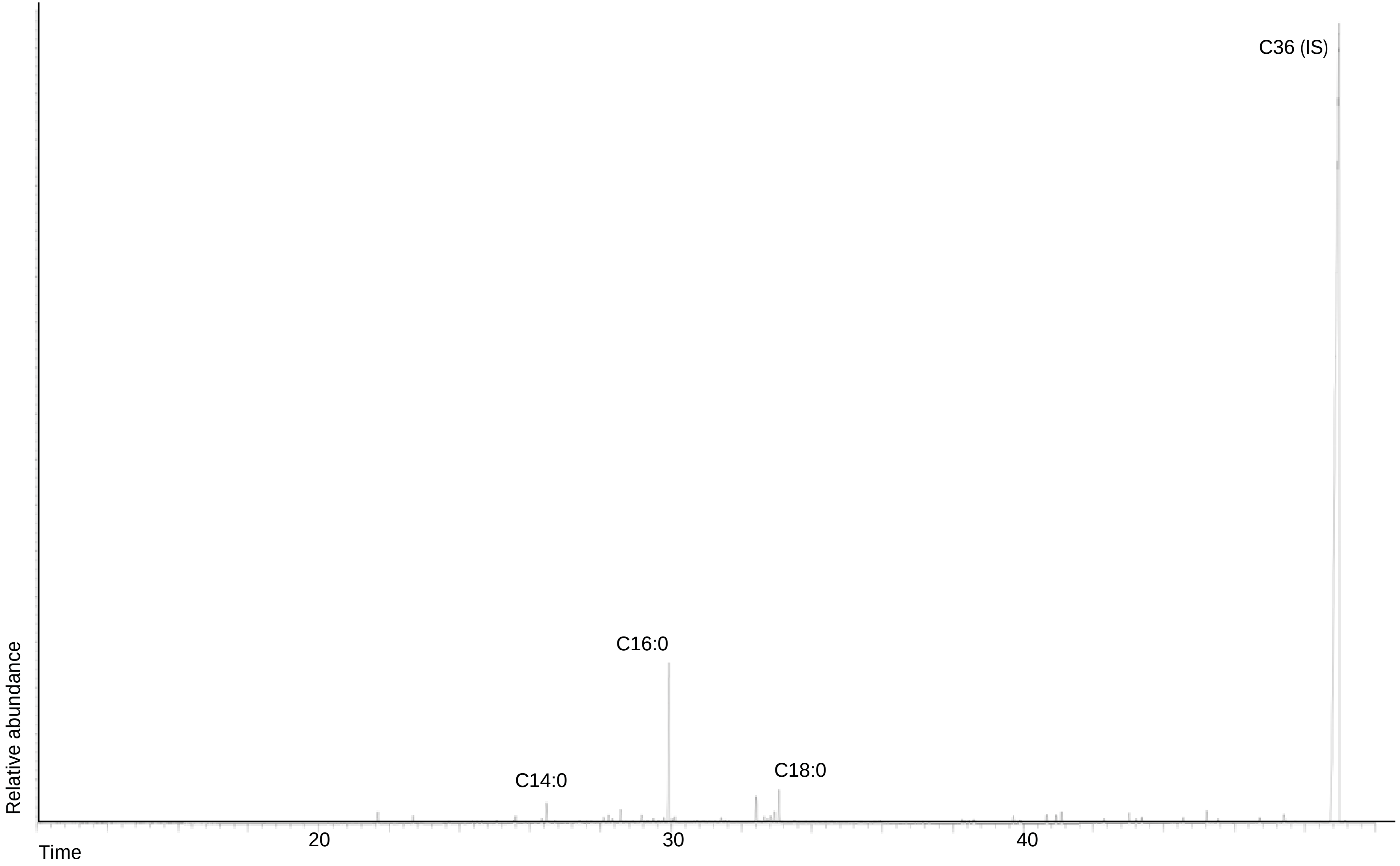
CV280



CV438

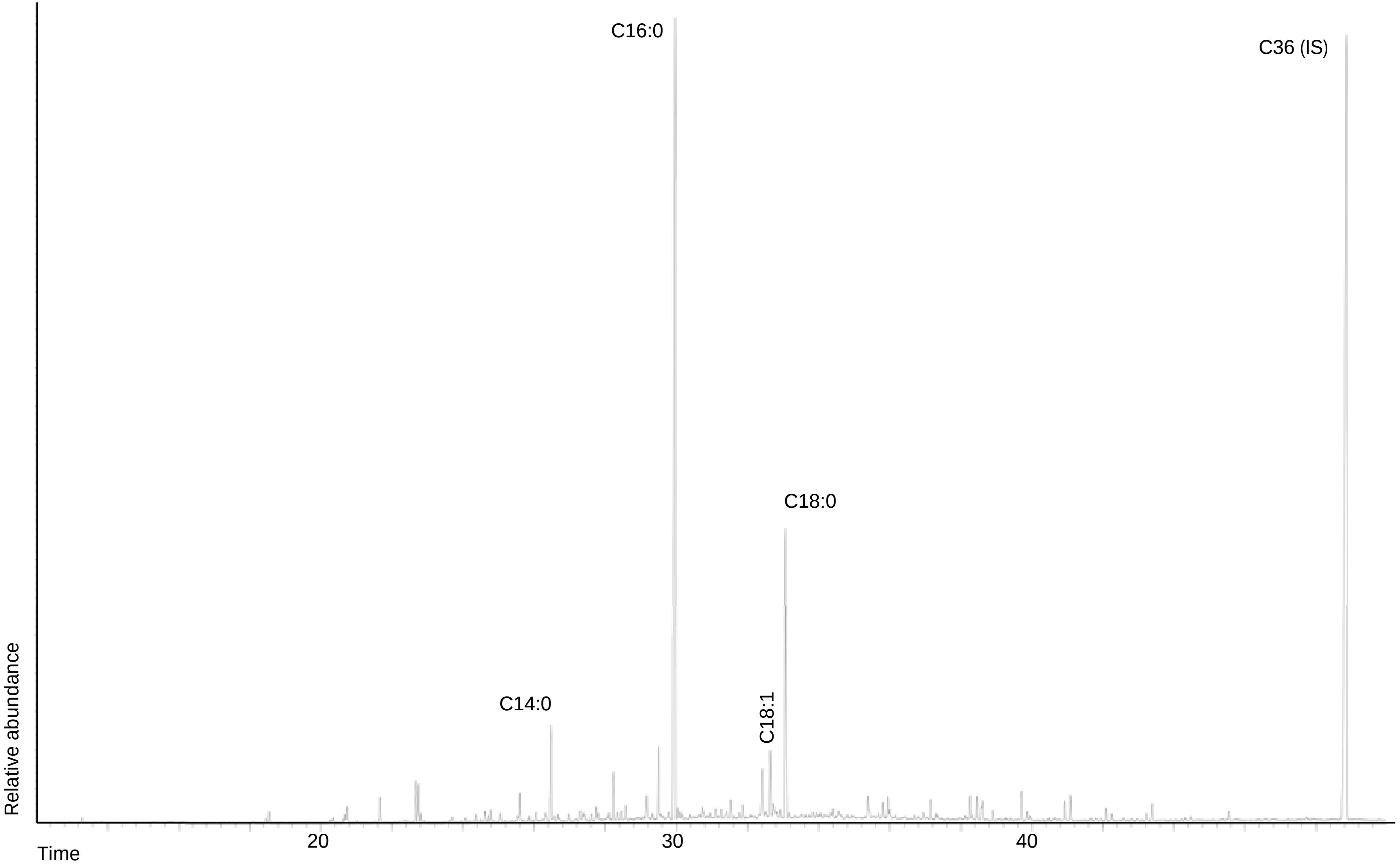


CV522*

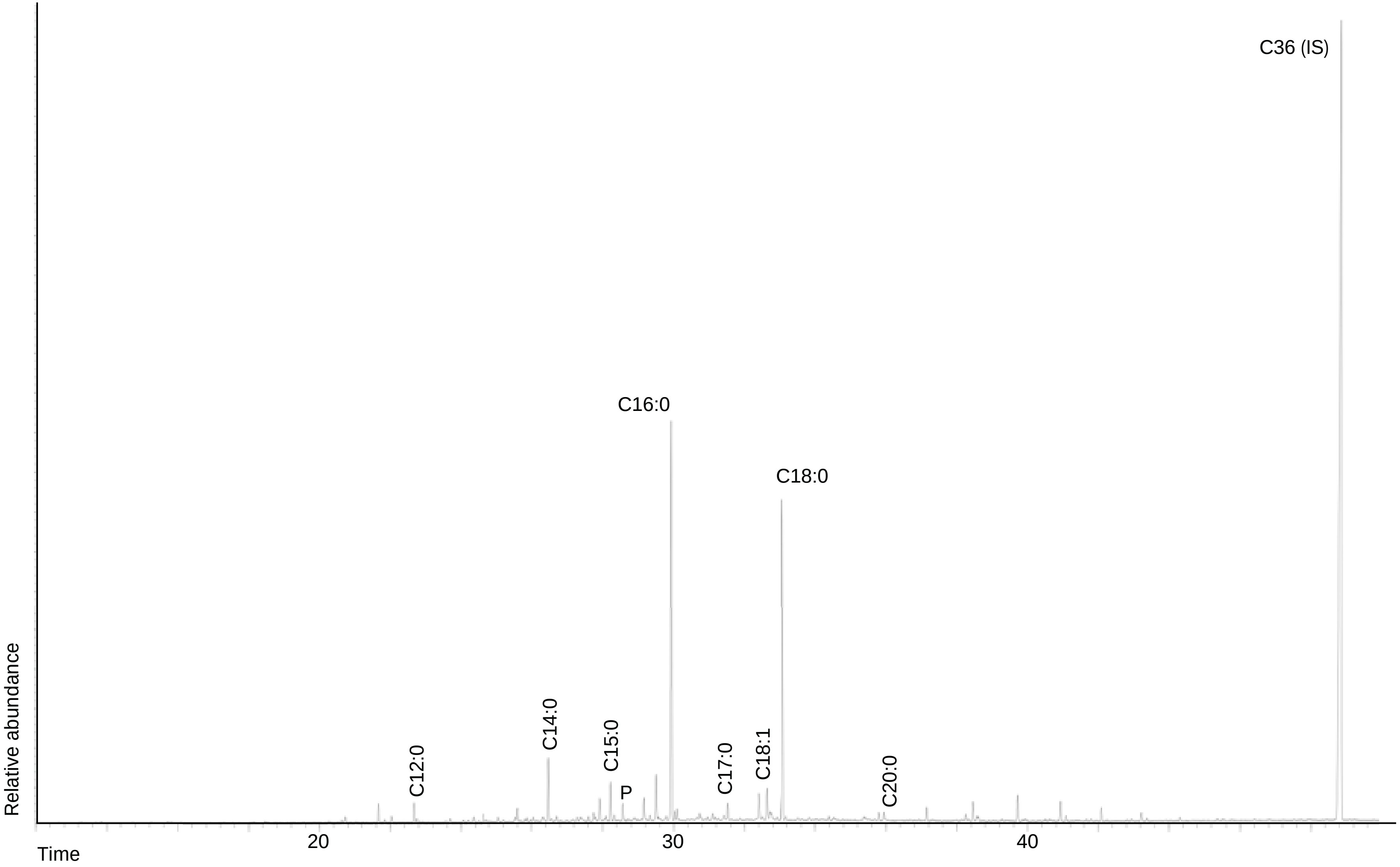


* this sample was diluted prior to the addition of the internal standard

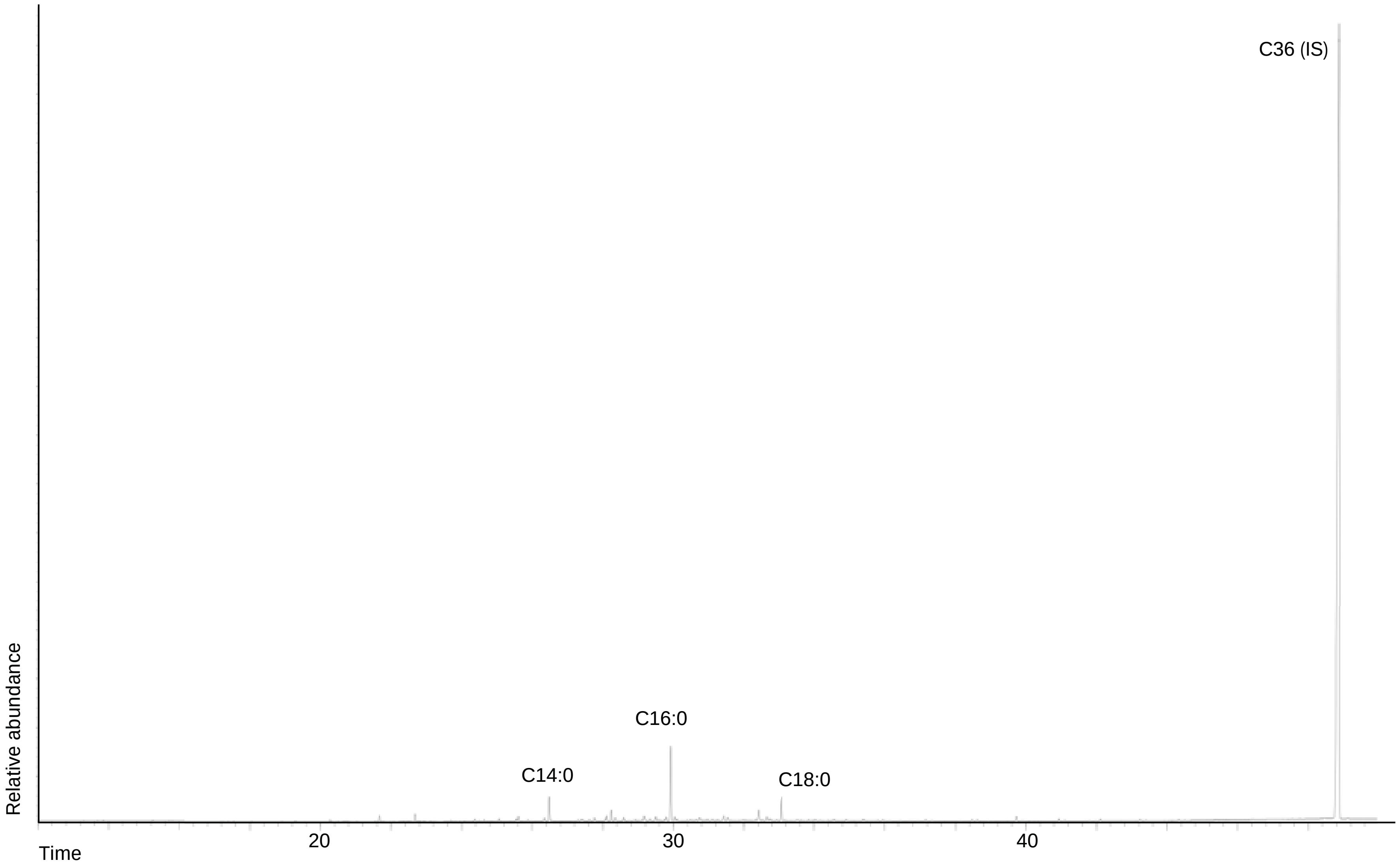
CV3003



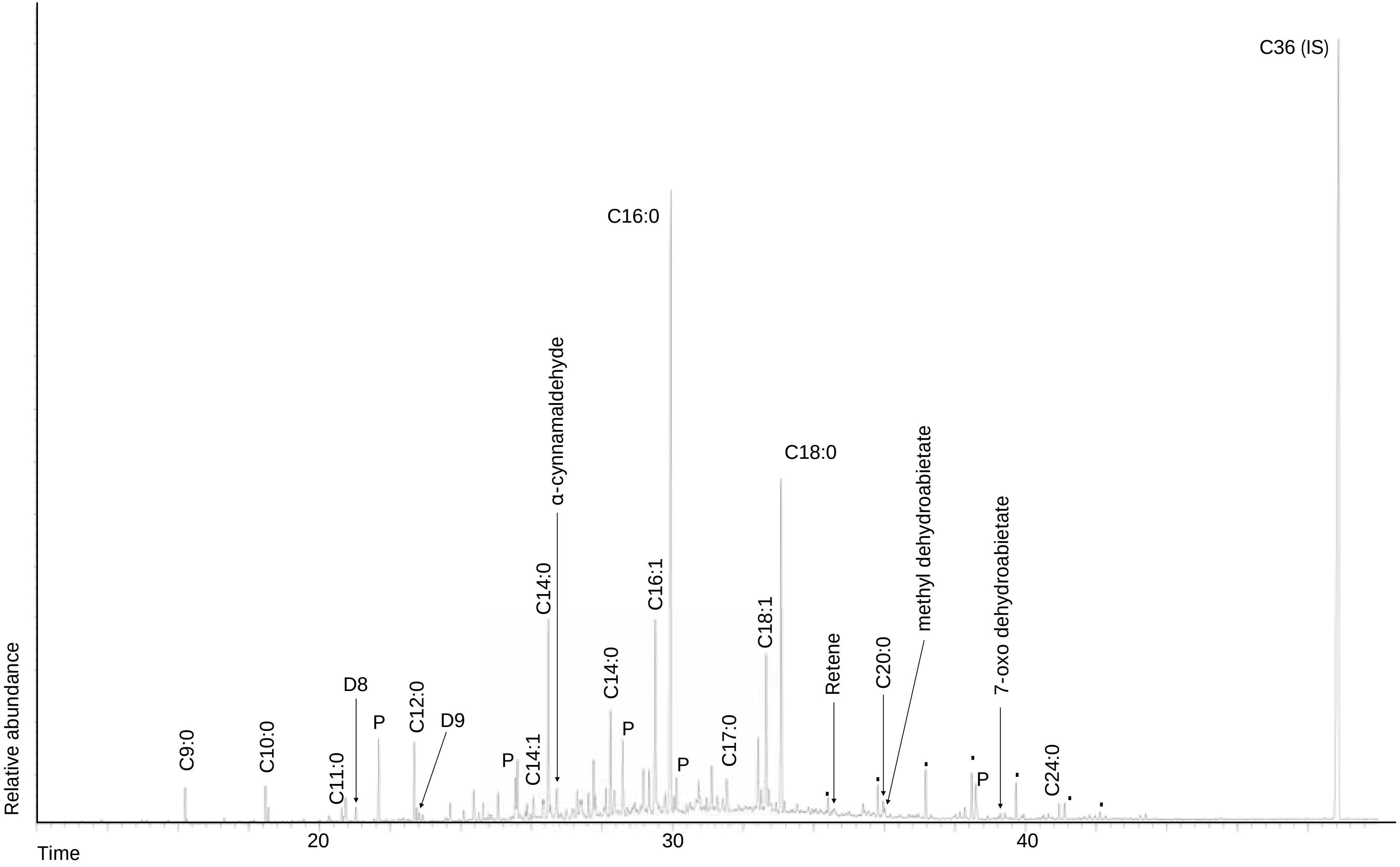
CVN56


C36 (IS)
C16:0
C18:0
C12:0
C14:0
C15:0
P
C17:0
C18:1
C20:0
Relative abundance
Time
20
30
40

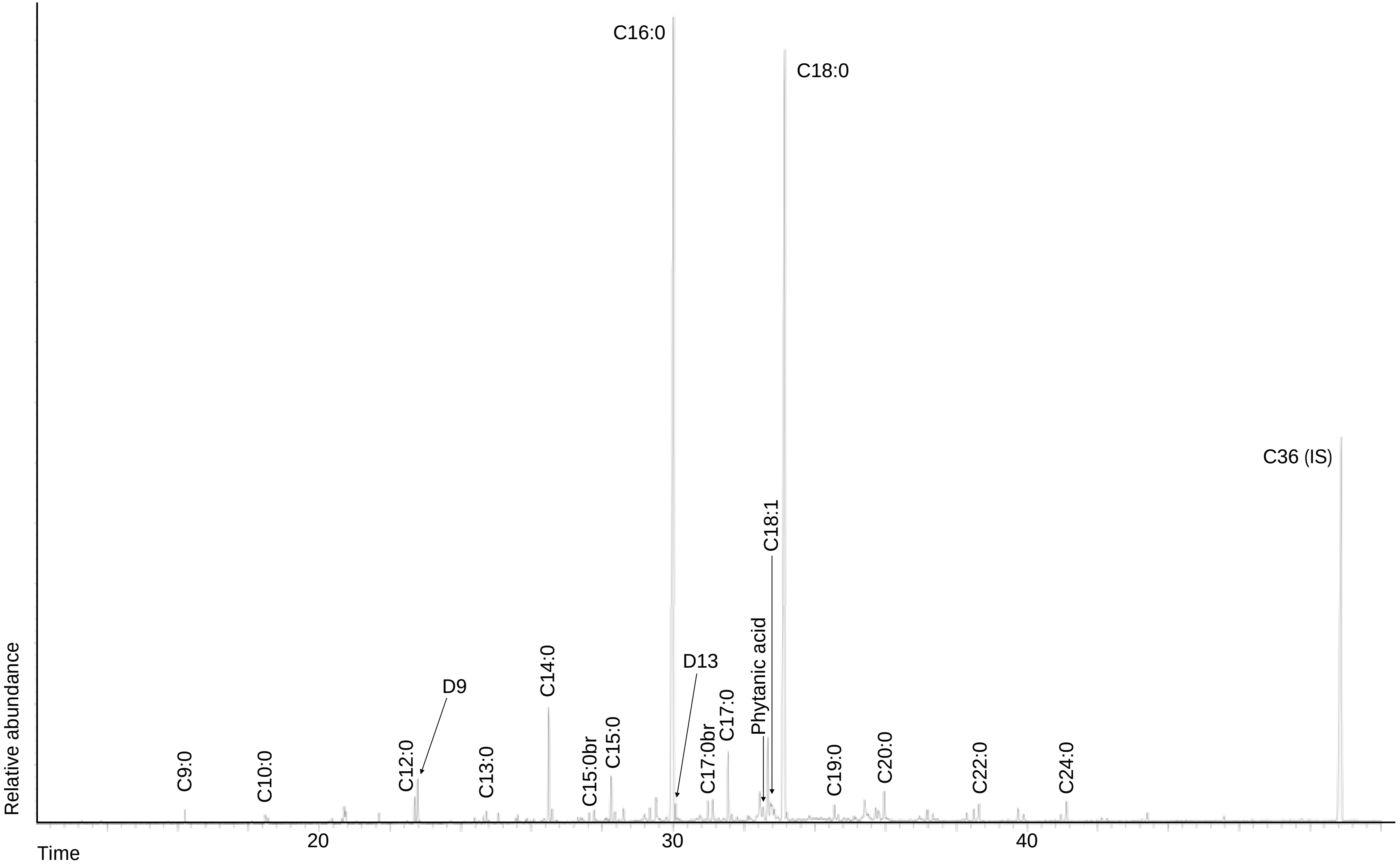
CVP1



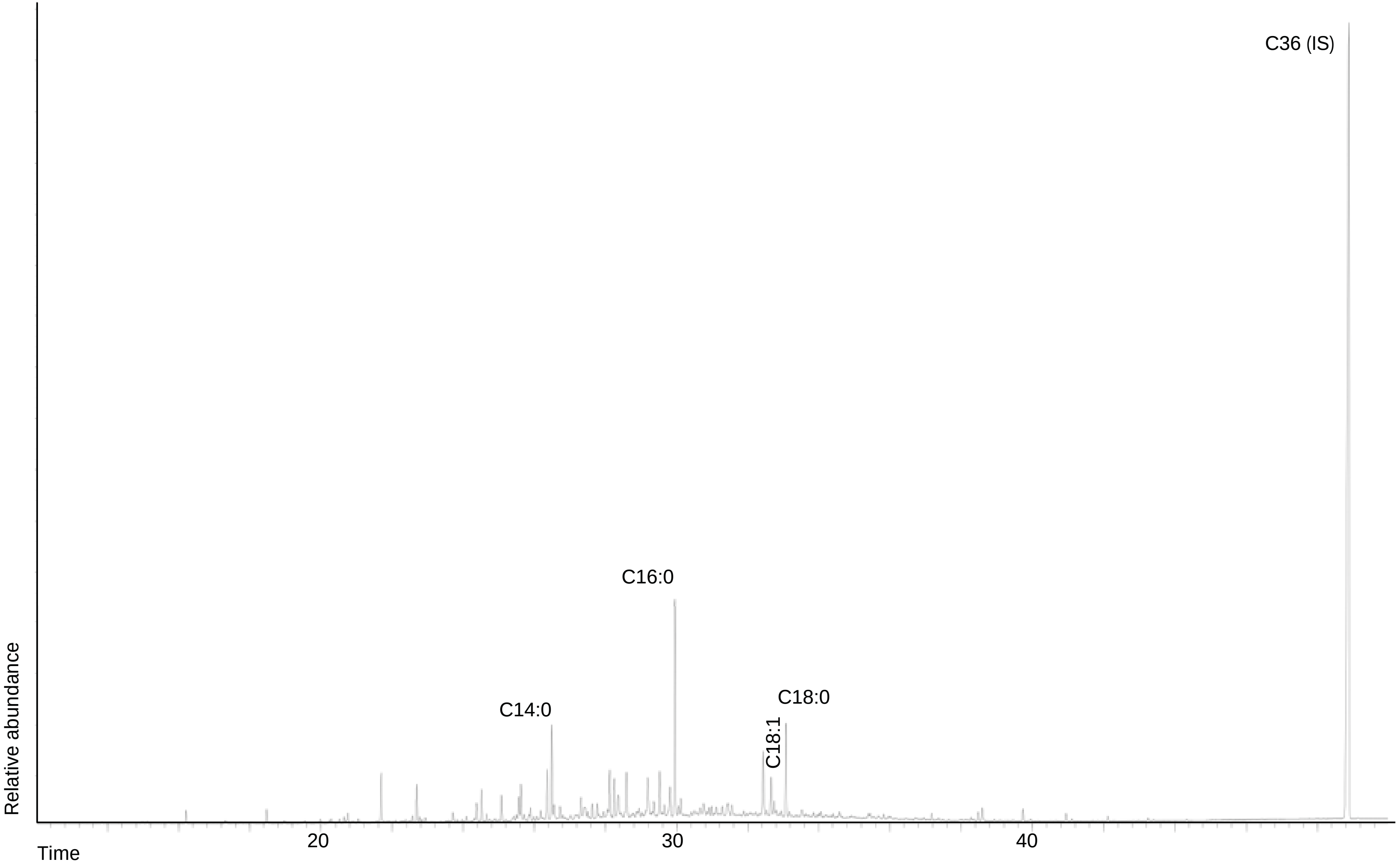
CVP2


C36 (IS)
C16:0
C18:0
α-cynnamaldehyde
C14:0
C16:1
C18:1
methyl dehydroabietate
7-oxo dehydroabietate
D8
P
C12:0
D9
C14:0
P
Retene
C20:0
C9:0
C10:0
C11:0
P
C14:1
P
C17:0
P
C24:0
Relative abundance
Time
20
30
40

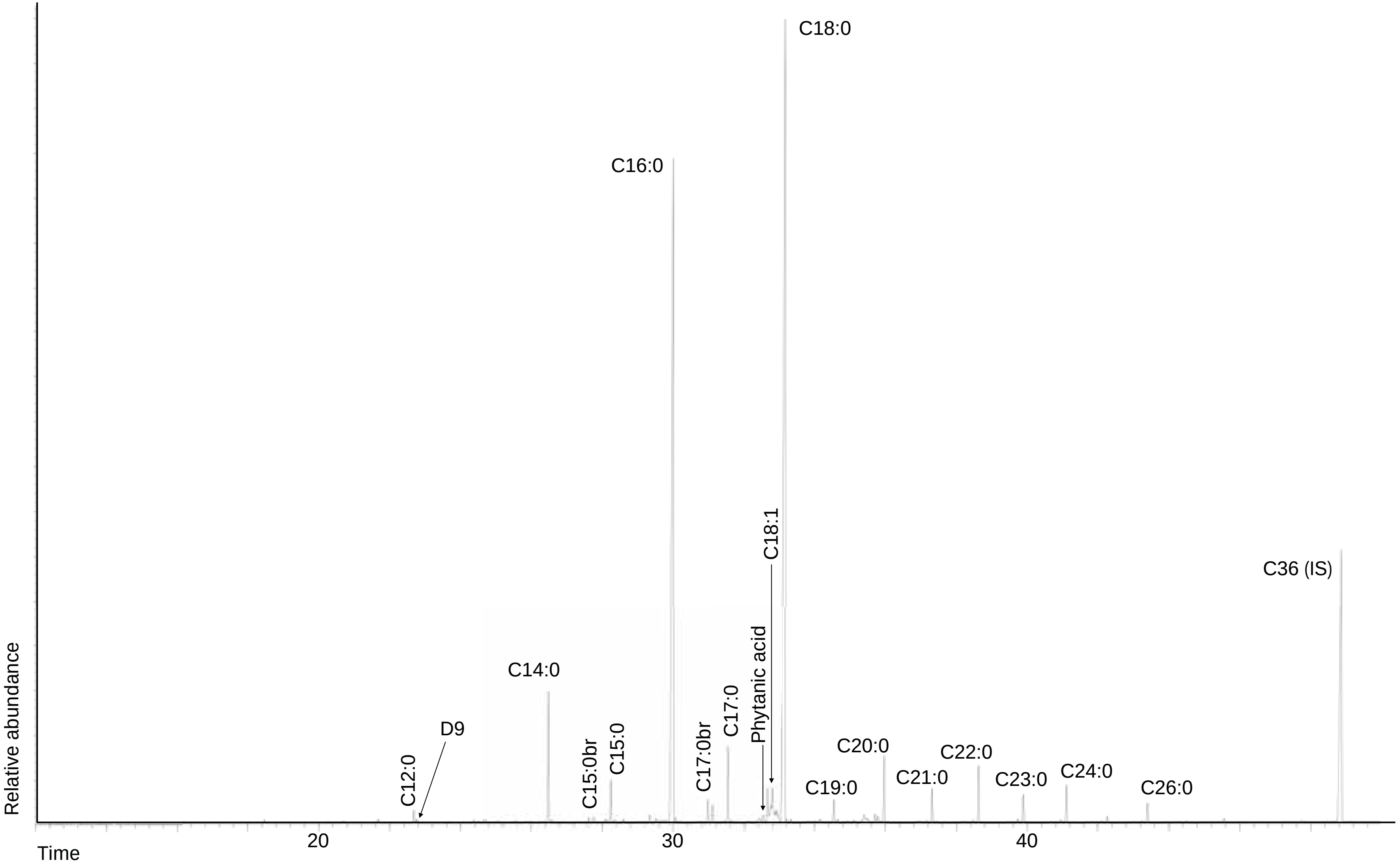
CVP3


C16:0
C18:0
C36 (IS)
C18:1
Phytanic acid
D13
C17:0
C17:0br
D9
C14:0
C15:0br
C15:0
C12:0
C13:0
C9:0
C10:0
C19:0
C20:0
C22:0
C24:0
Relative abundance
Time
20
30
40

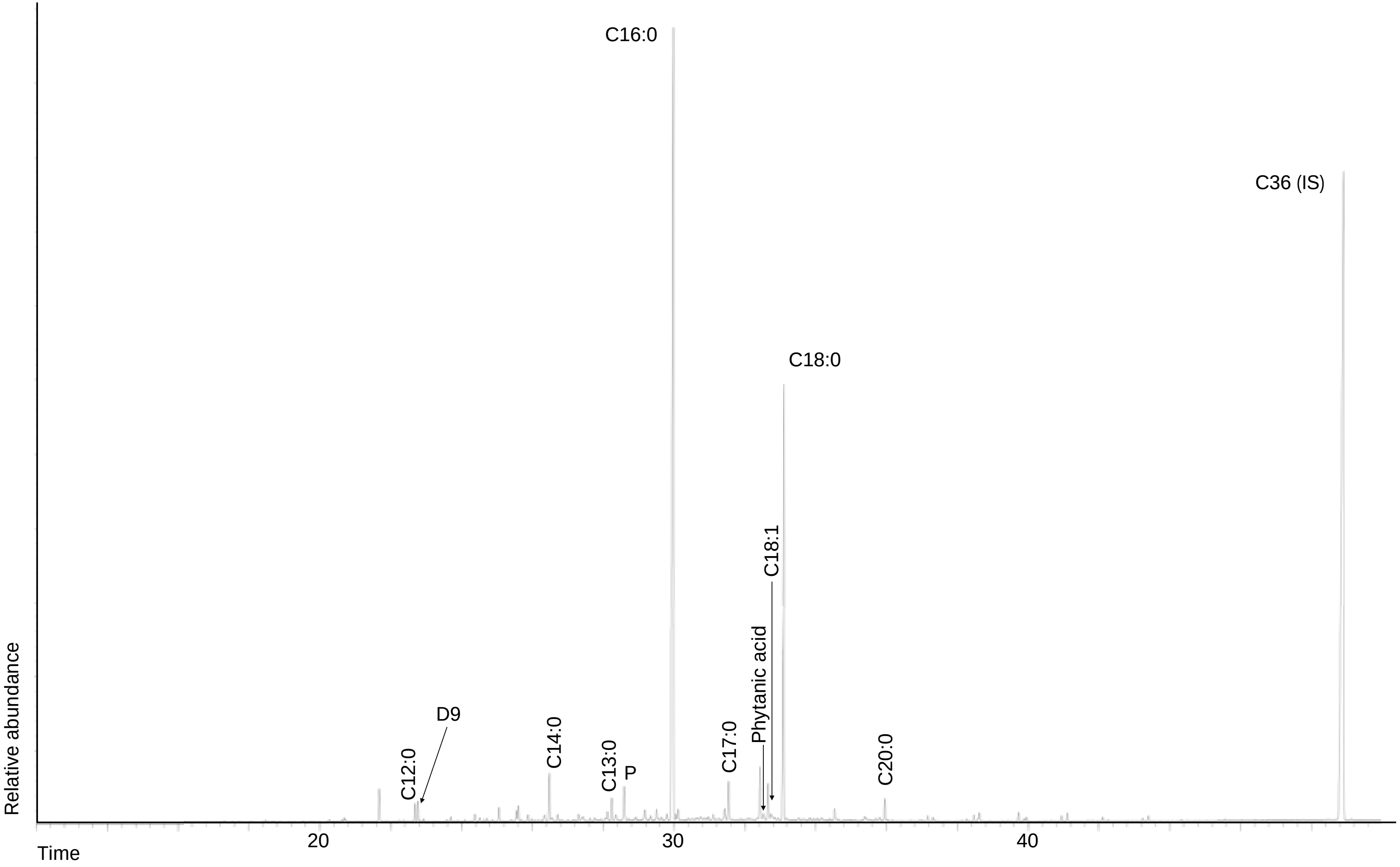
CVP4



CVP5

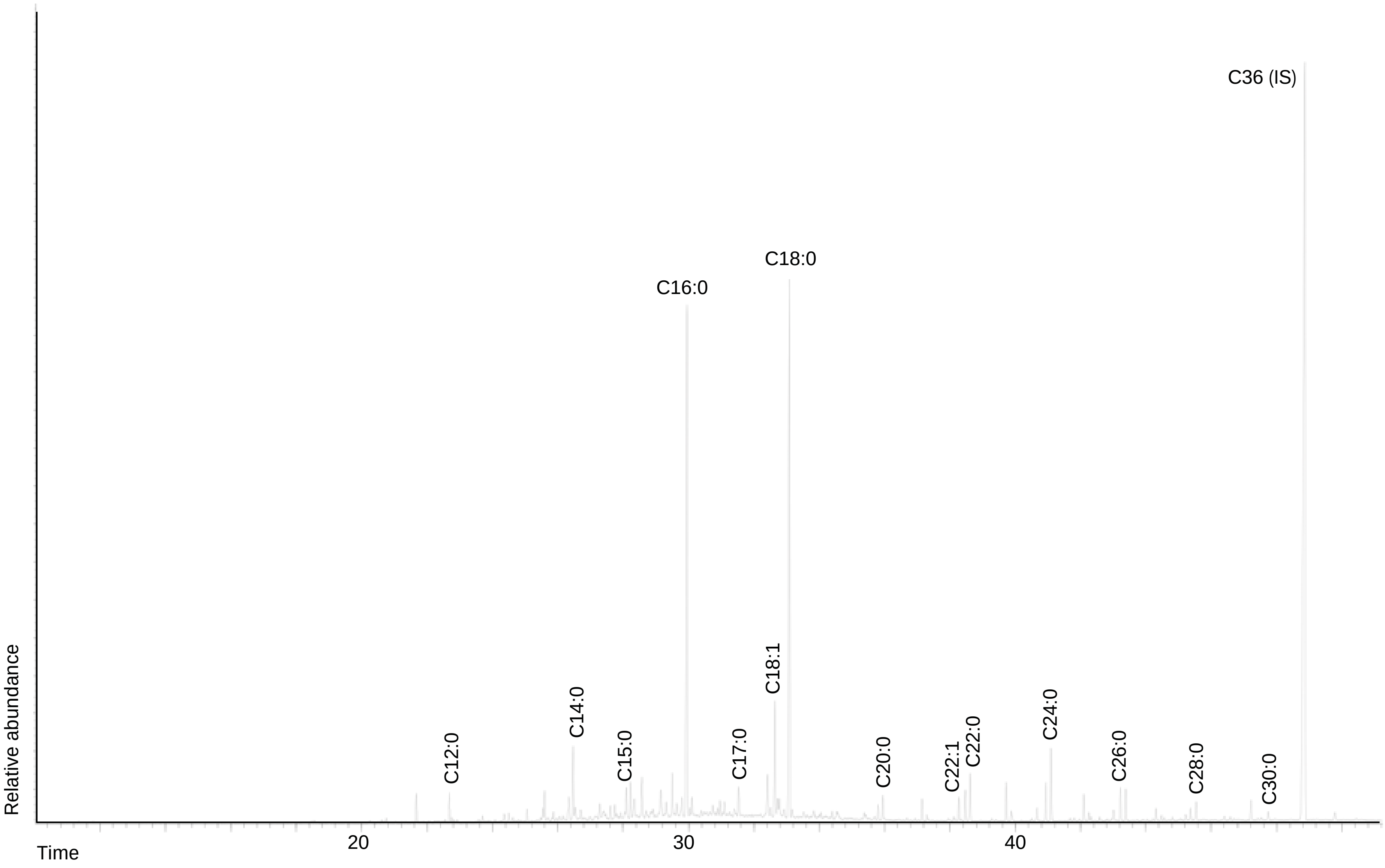

C18:0
C16:0
C18:1
C36 (IS)
C14:0
Phytanic acid
C17:0
D9
C17:0br
C15:0
C15:0br
C20:0
C12:0
C22:0
C21:0
C24:0
C19:0
C23:0
C26:0
Relative abundance
Time
20
30
40

CVP6

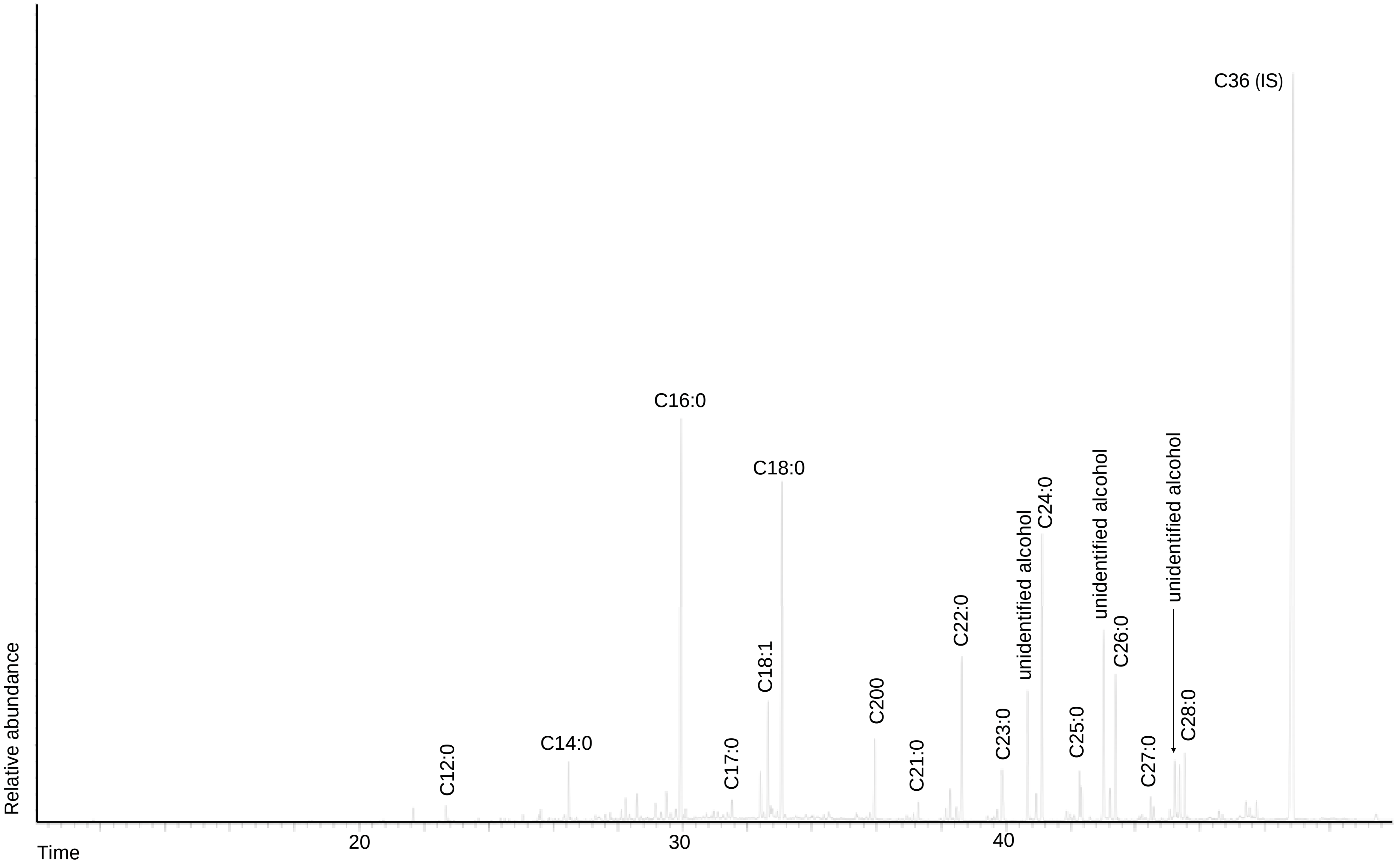

C16:0
C36 (IS)
C18:0
C18:1
Phytanic acid
D9
C12:0
C14:0
C13:0
P
C17:0
C20:0
Relative abundance
Time
20
30
40

Cova de la Font Major

FM7

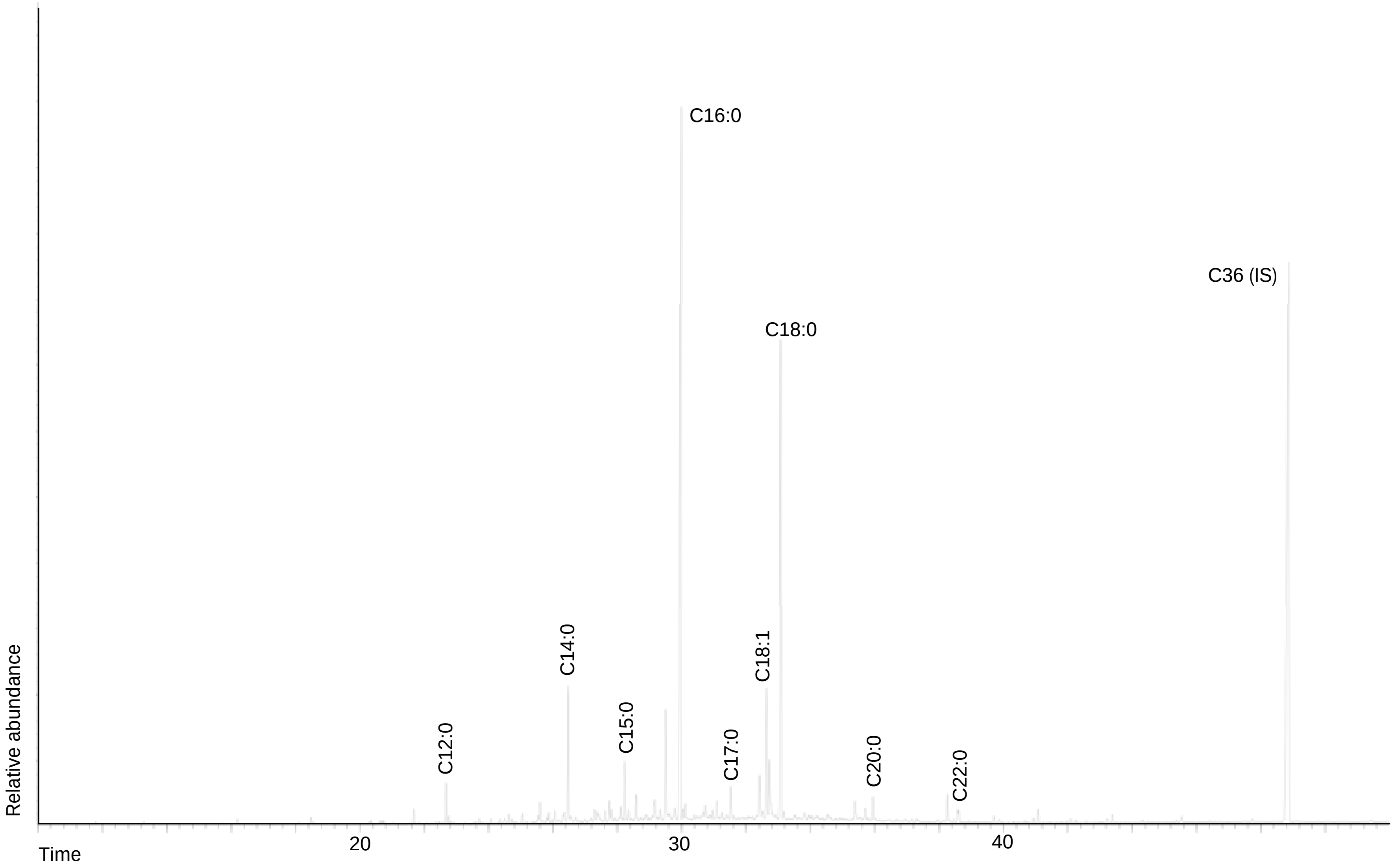

C36 (IS)
C18:0
C16:0
C18:1
C14:0
C24:0
C22:0
C12:0
C20:0
C26:0
C22:1
C15:0
C17:0
C28:0
C30:0
Relative abundance
Time
20
30
40

FM14



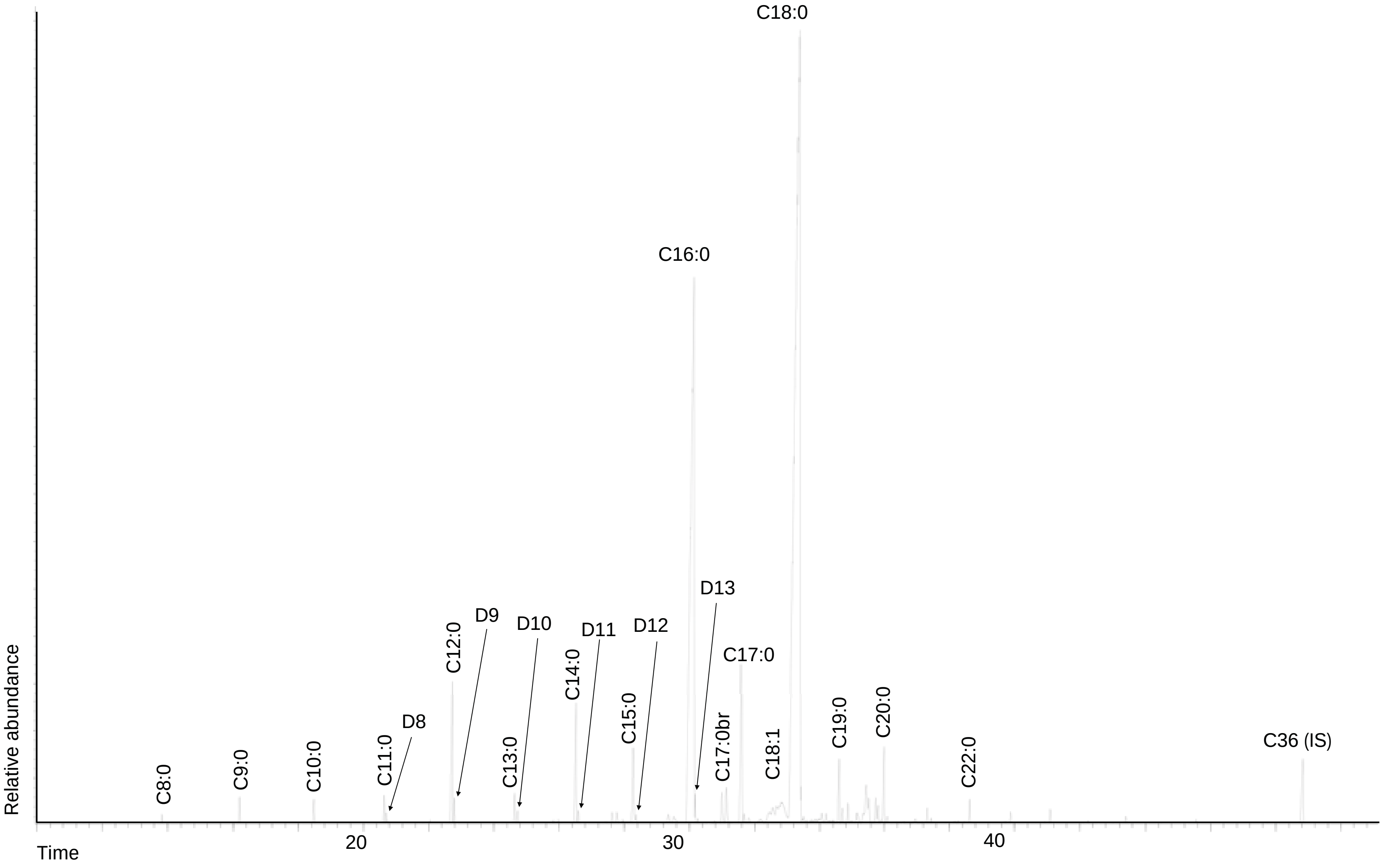
Relative abundance
Time
20
30
40
C12:0
C14:0
C16:0
C17:0
C18:1
C18:0
C200
C21:0
C22:0
C23:0
unidentified alcohol
C24:0
C25:0
unidentified alcohol
C26:0
C27:0
unidentified alcohol
C28:0
C36 (IS)

FM62

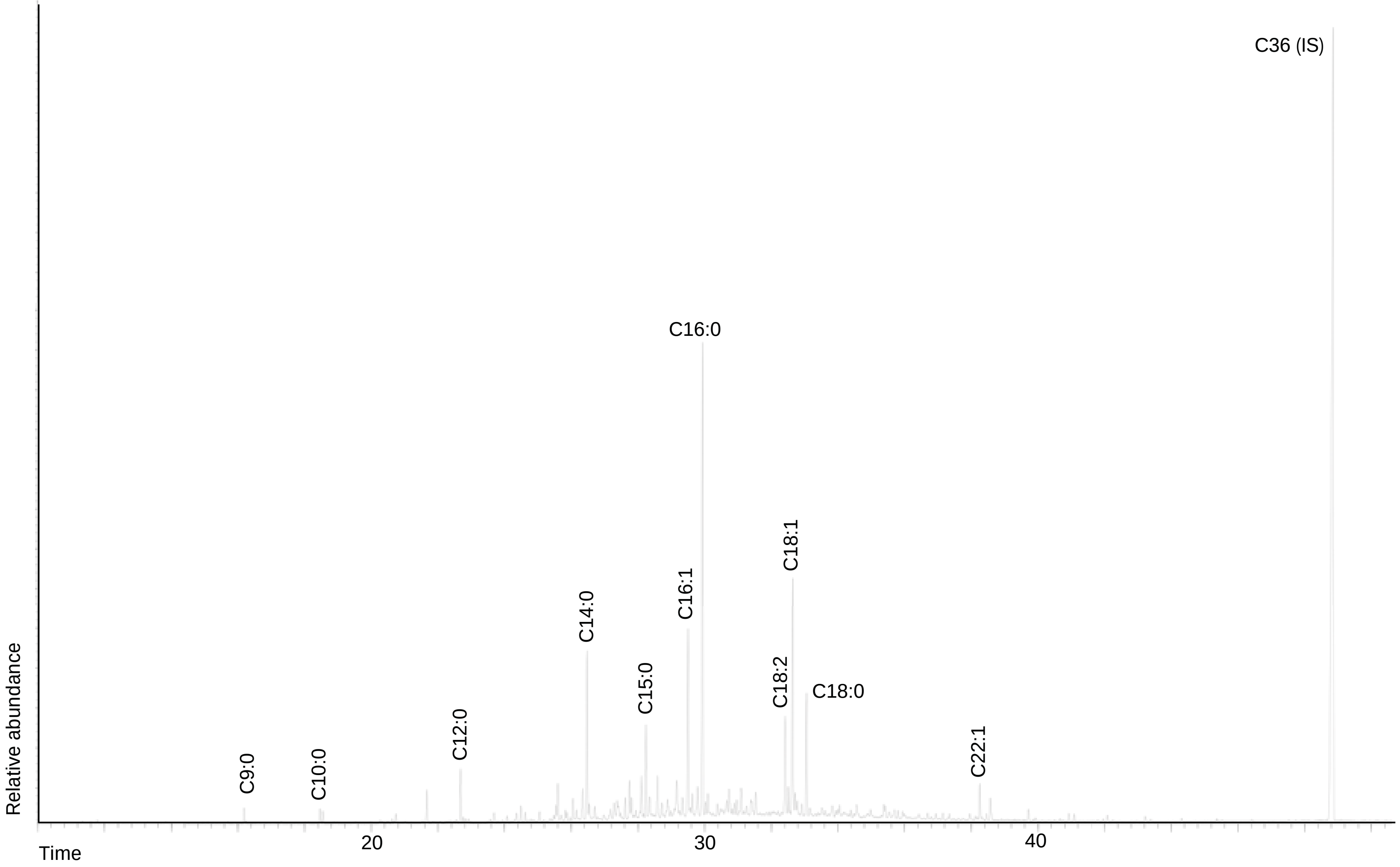


C16:0
C36 (IS)
C18:0
C14:0
C18:1
C15:0
C12:0
C17:0
C20:0
C22:0
Relative abundance
Time
20
30
40

FM81

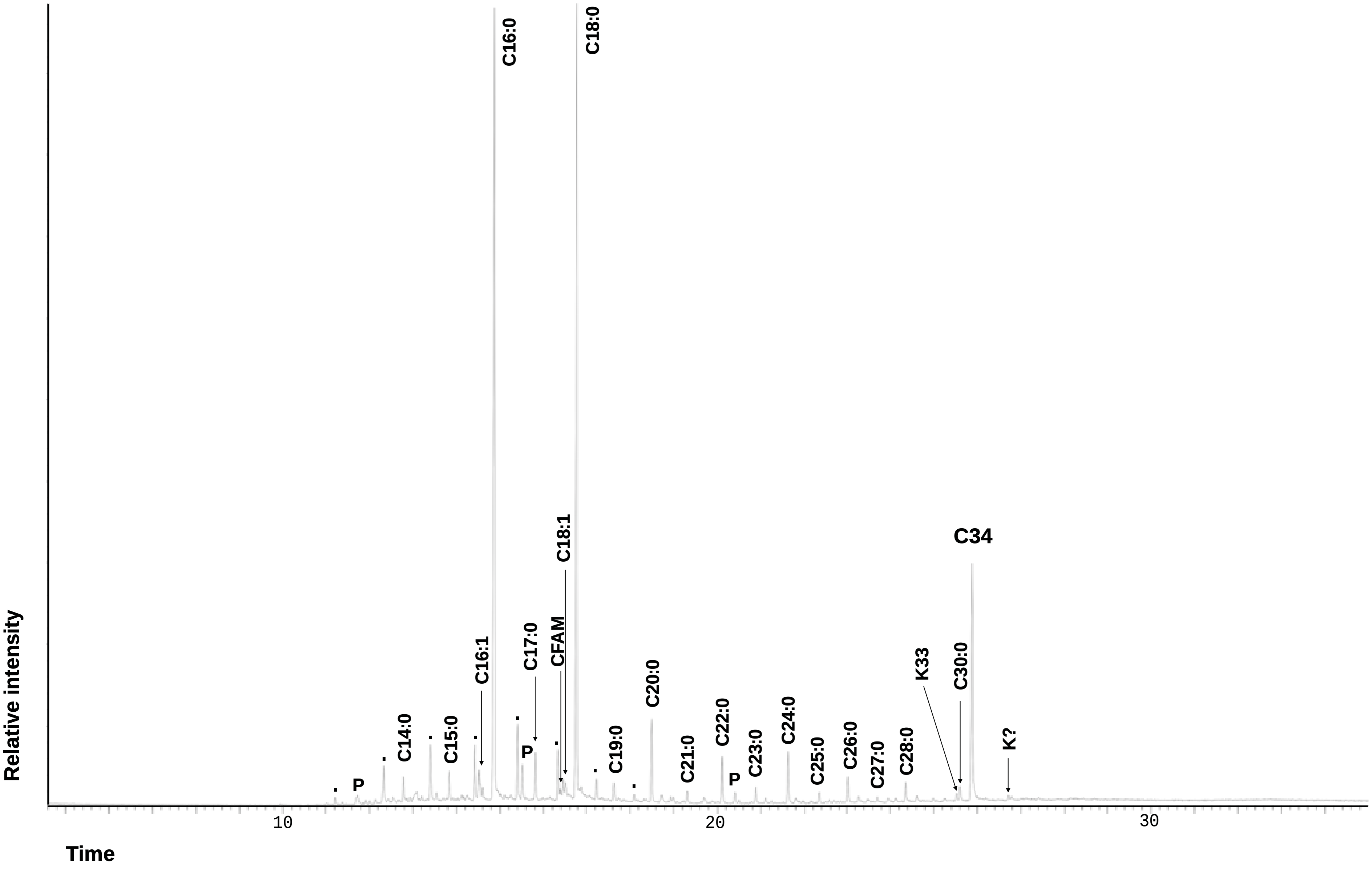

C18:0
C16:0
D13
D9
D10
D11
D12
C17:0
C12:0
C14:0
D8
C15:0
C17:0br
C20:0
C19:0
C36 (IS)
C18:1
C8:0
C9:0
C10:0
C11:0
C13:0
C22:0
Relative abundance
Time
20
30
40

FM84

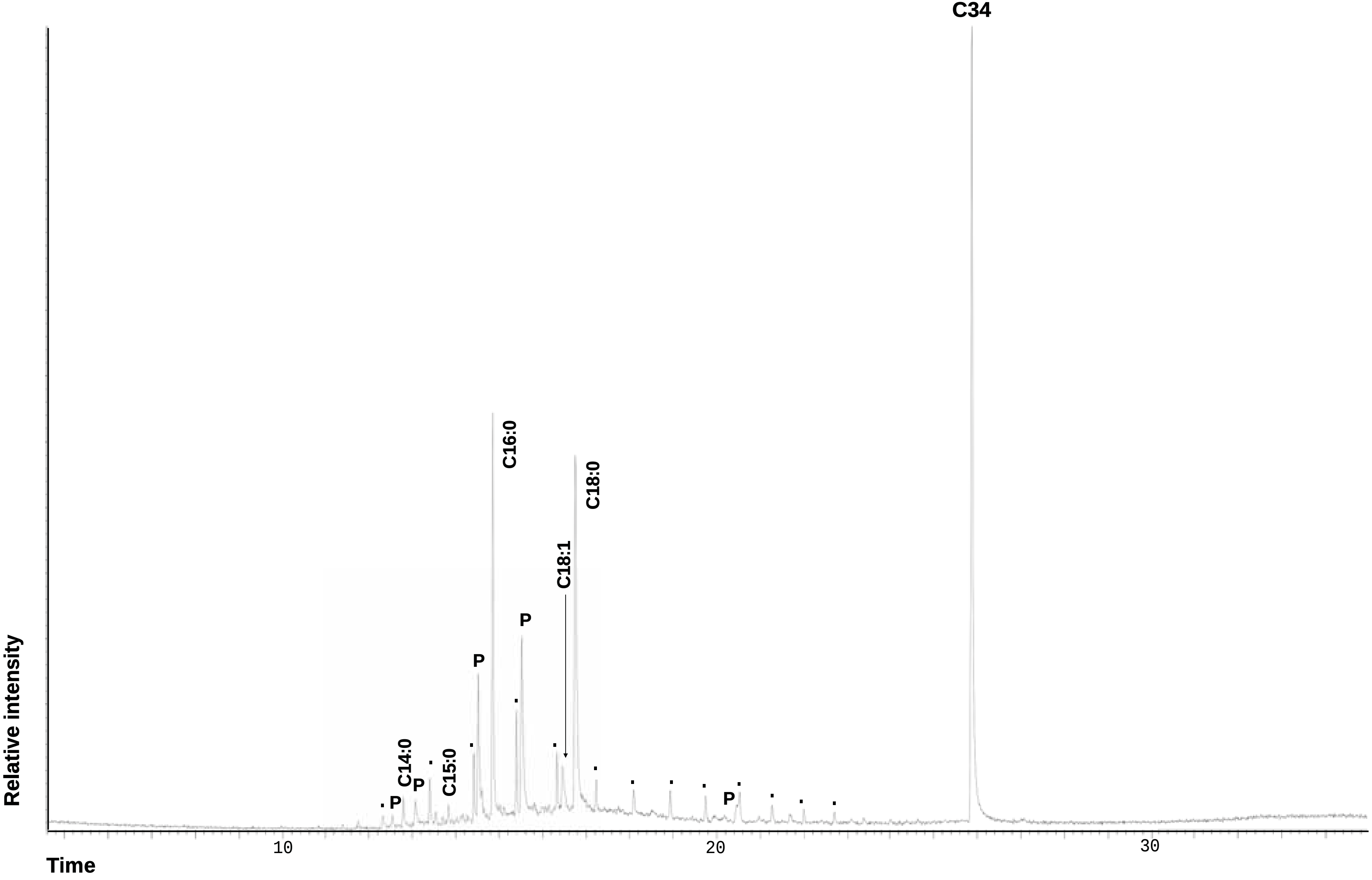

C36 (IS)
C16:0
C18:1
C16:1
C14:0
C15:0
C18:2
C18:0
C12:0
C22:1
C10:0
C9:0
Relative abundance
Time
20
30
40

Cova del Vidre

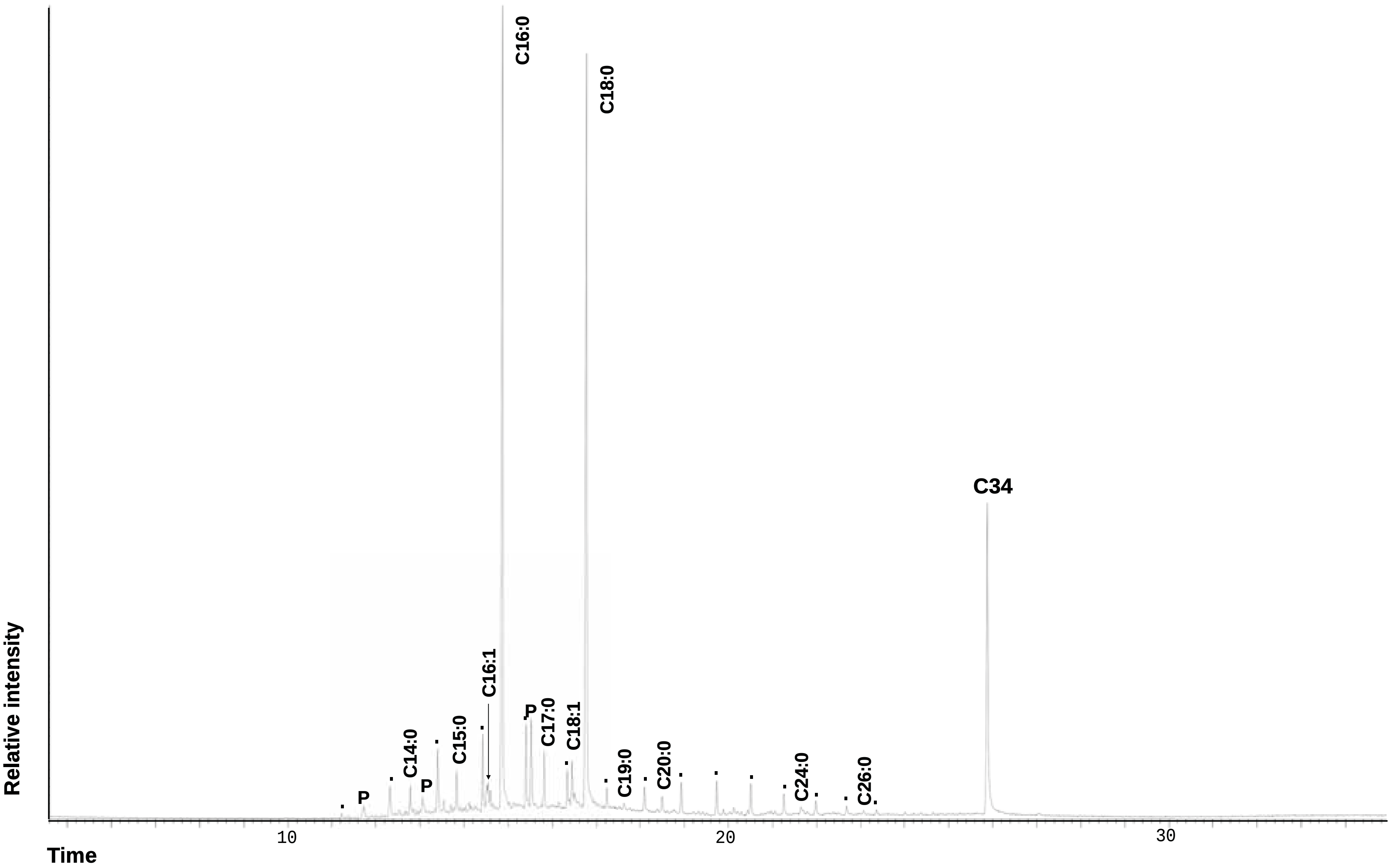
Vid107


Relative intensity
P
C14:0
C15:0
C16:1
C16:0
P
C17:0
CFAM
C18:1
C18:0
C19:0
C20:0
C21:0
C22:0
P
C23:0
C24:0
C25:0
C26:0
C27:0
C28:0
K33
C30:0
C34
K?
10
20
30
Time

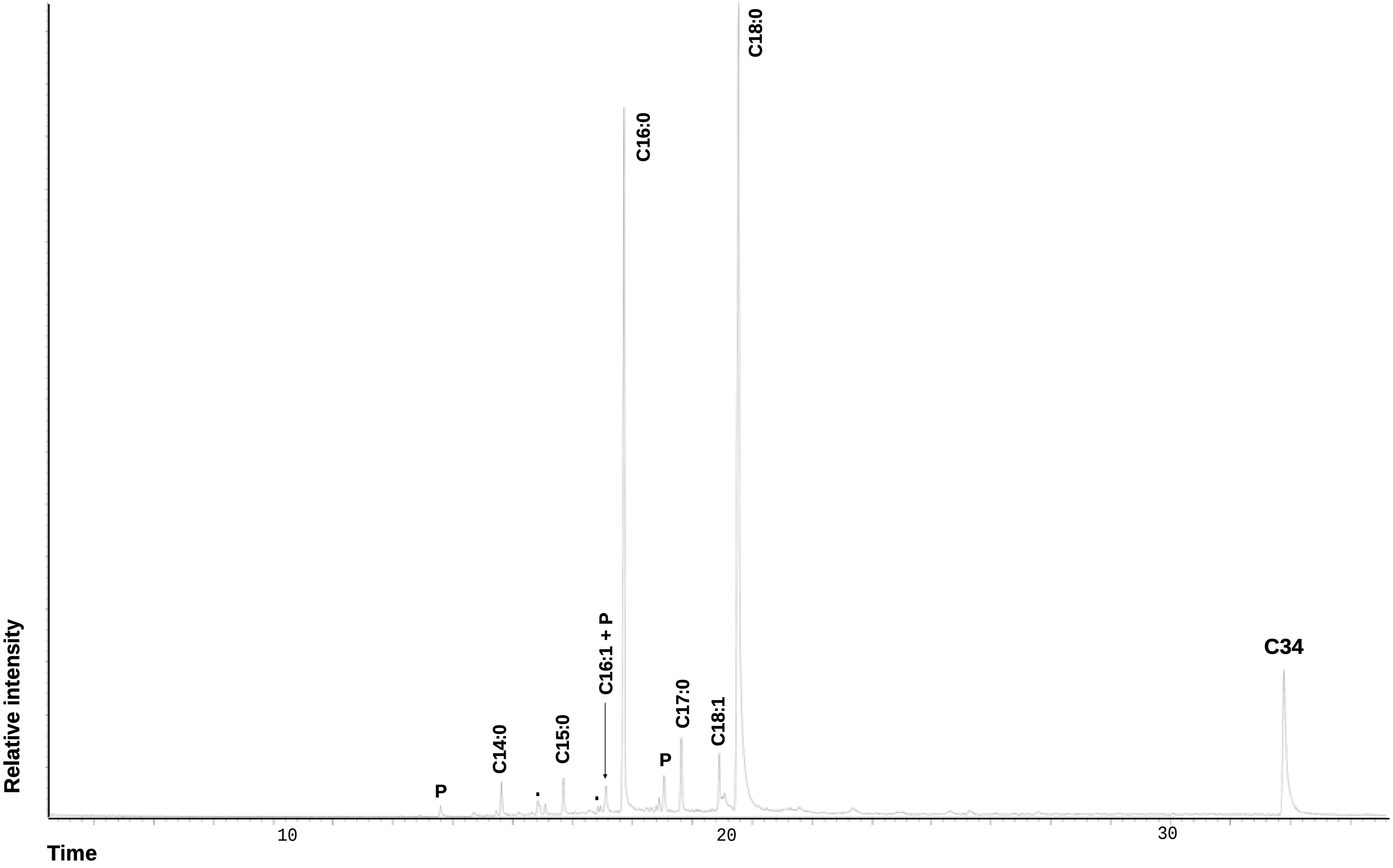
Vid 28


C34
C16:0
C18:0
C18:1
P
P
P
P
P
P
C14:0
C15:0
Relative intensity
Time
10
20
30

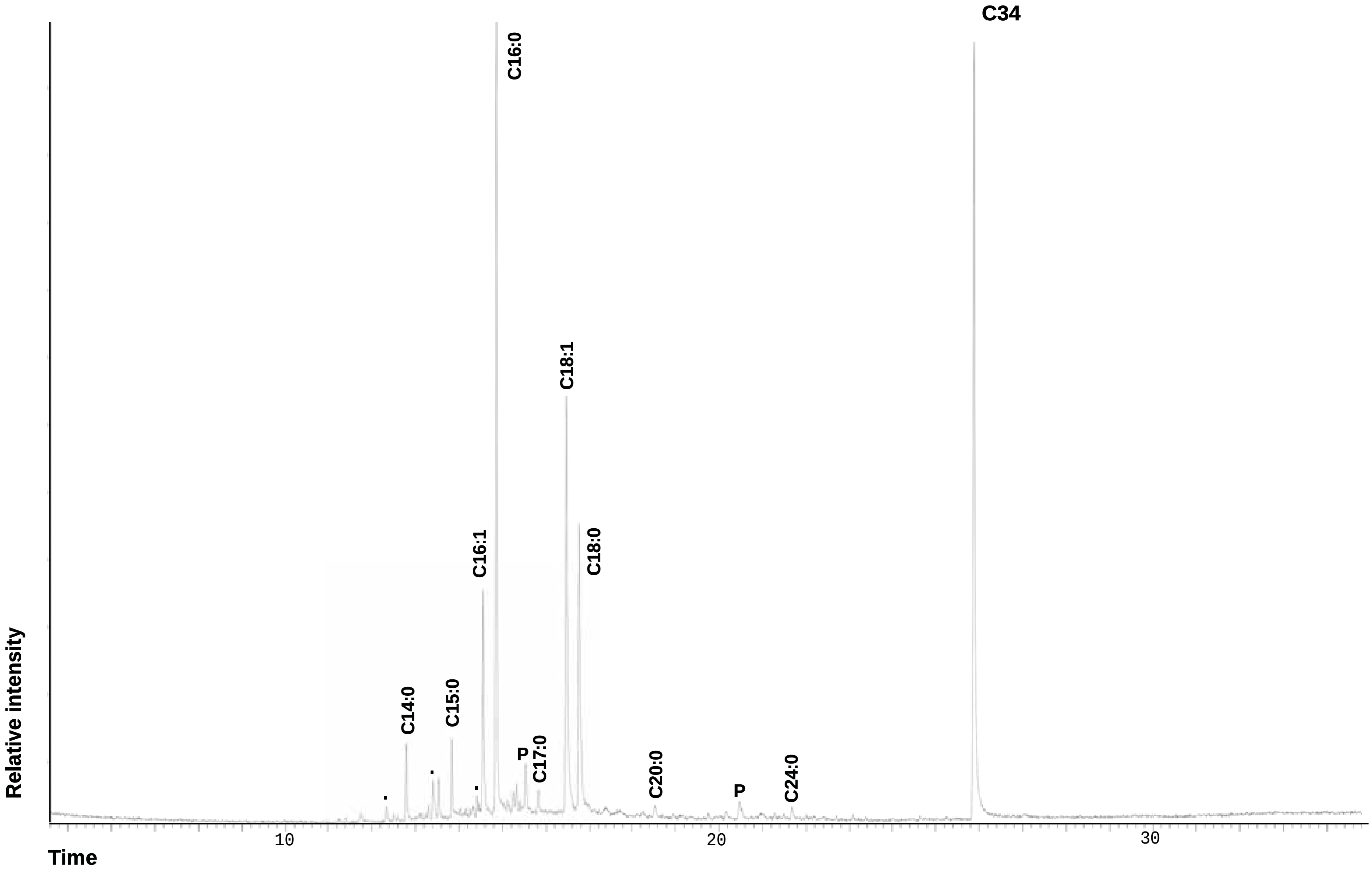
Vid 117


C16:0
C18:0
C34
P
C14:0
P
C15:0
C16:1
P
C17:0
C18:1
C19:0
C20:0
C24:0
C26:0
Relative intensity
10
20
30
Time

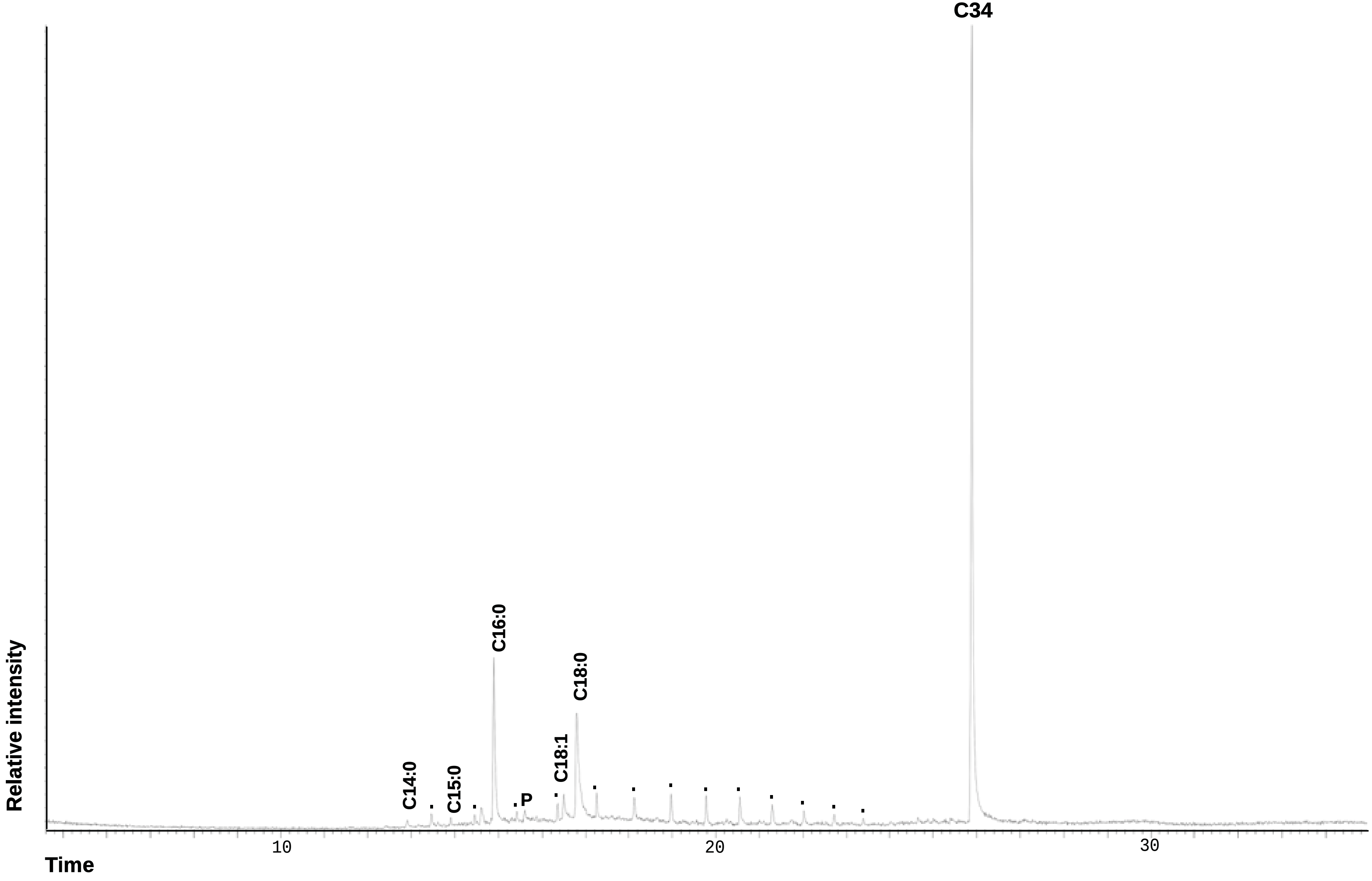
Vid 44


C18:0
C16:0
C34
C16:1 + P
C17:0
C18:1
C15:0
C14:0
P
P
Relative intensity
10
20
30
Time

Vid 90


C34
C16:0
C18:1
C18:0
C16:1
C14:0
C15:0
P
C17:0
C20:0
P
C24:0
Relative intensity
10
20
30
Time

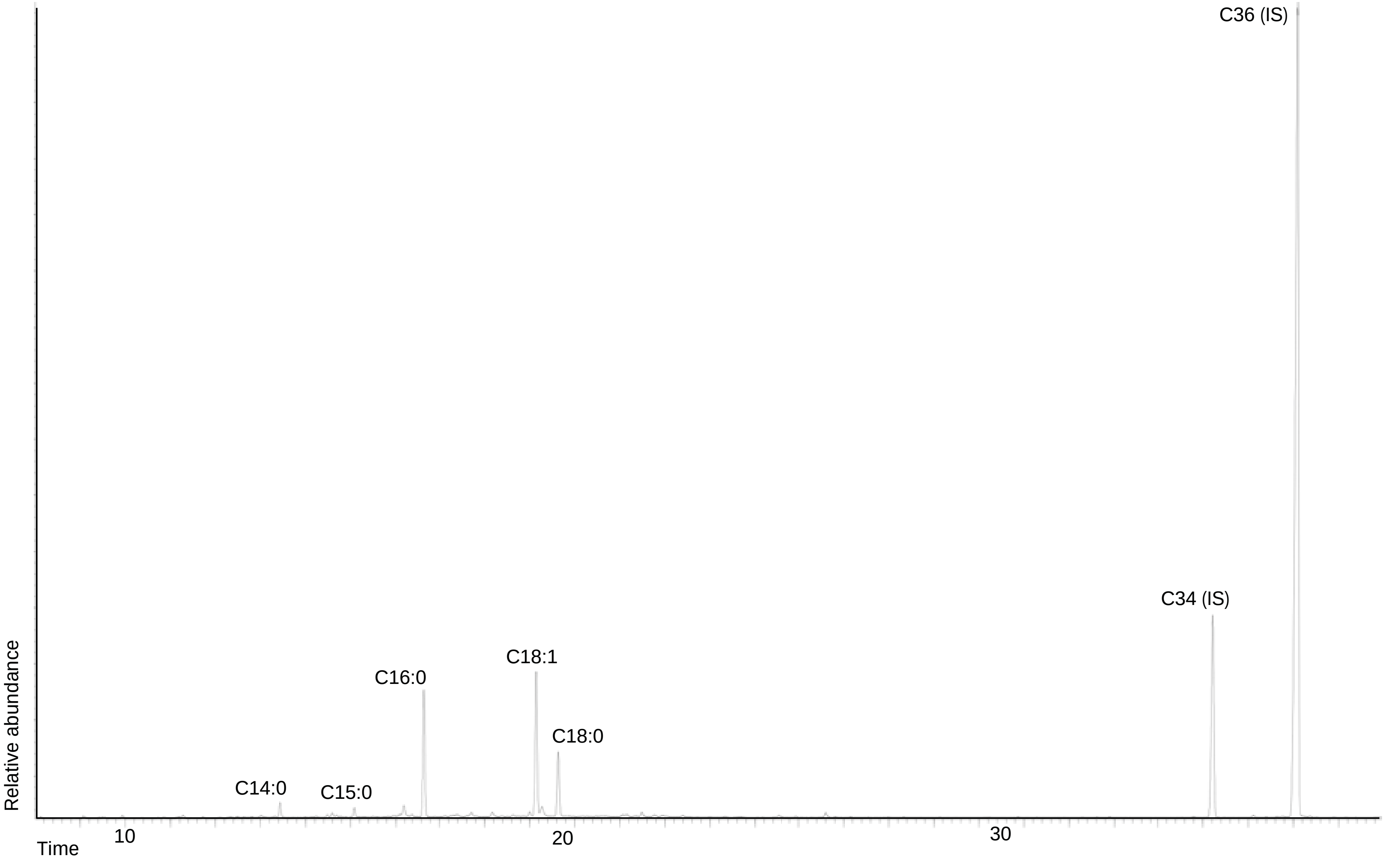
Vid 1047



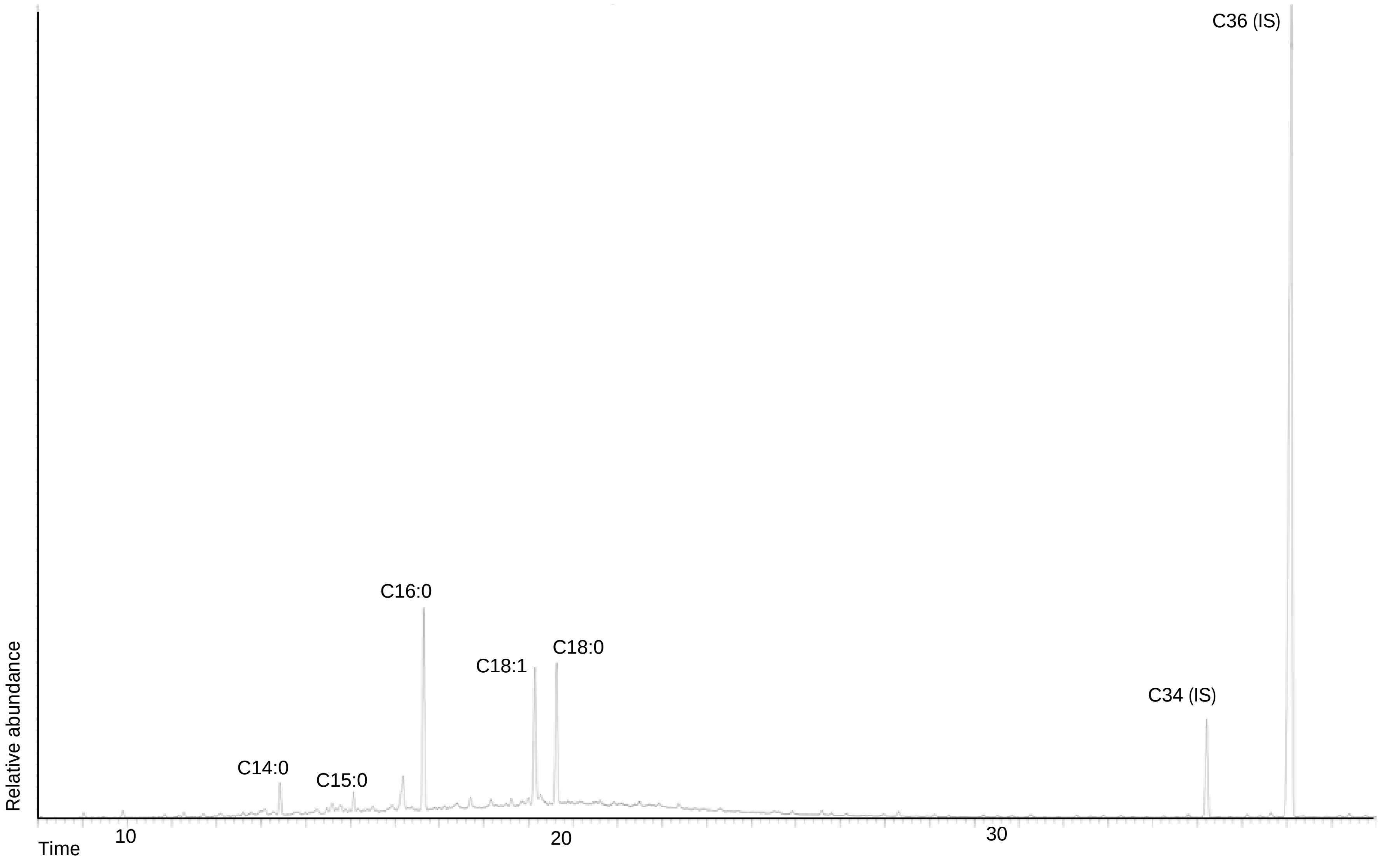
C34
C16:0
C18:0
C18:1
C14:0
C15:0
P
Relative intensity
Time
10
20
30

Barranc d’en Fabra

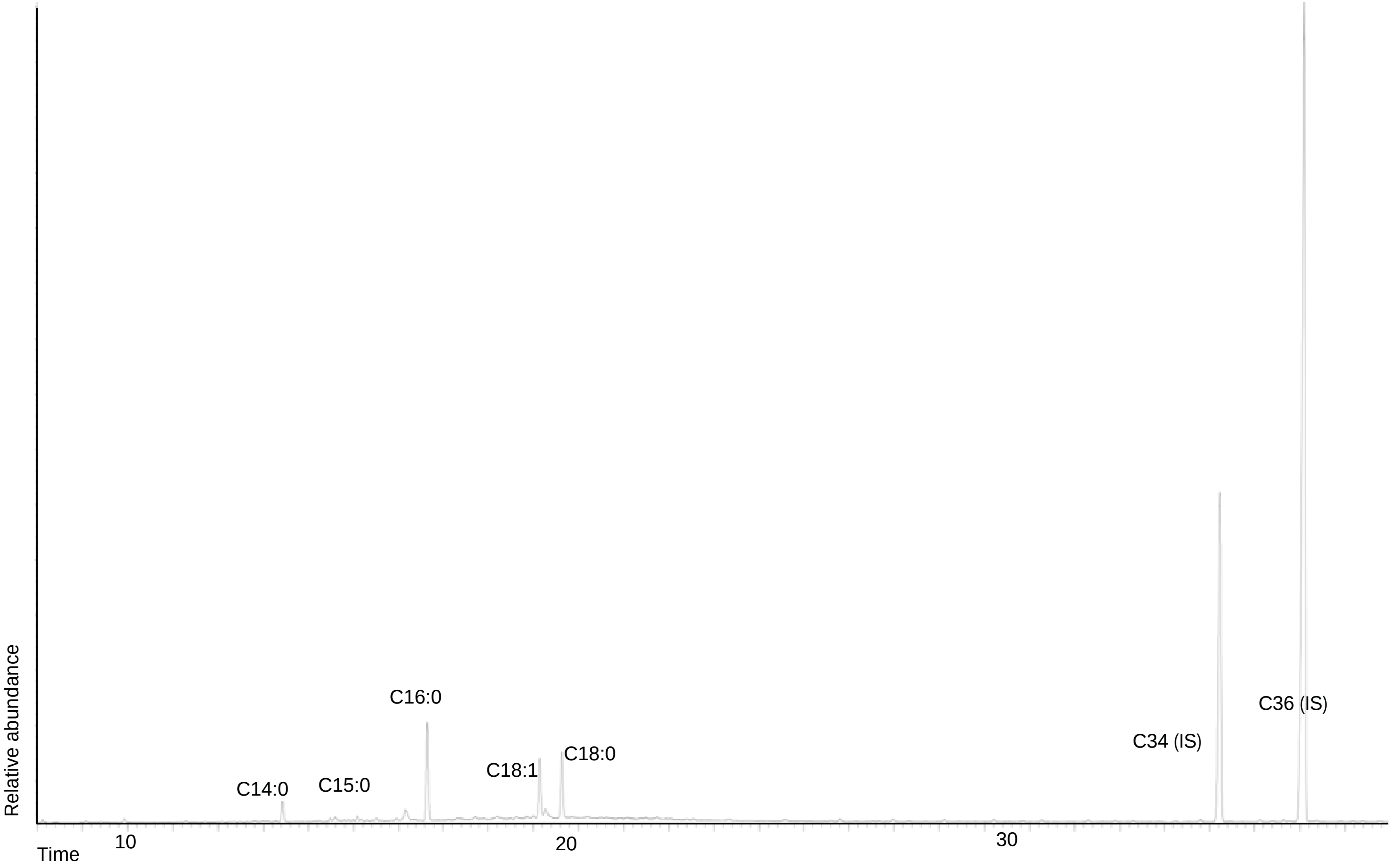
BF1


C36 (IS)
C34 (IS)
C18:1
C16:0
C18:0
C14:0
C15:0
Relative abundance
Time
10
20
30

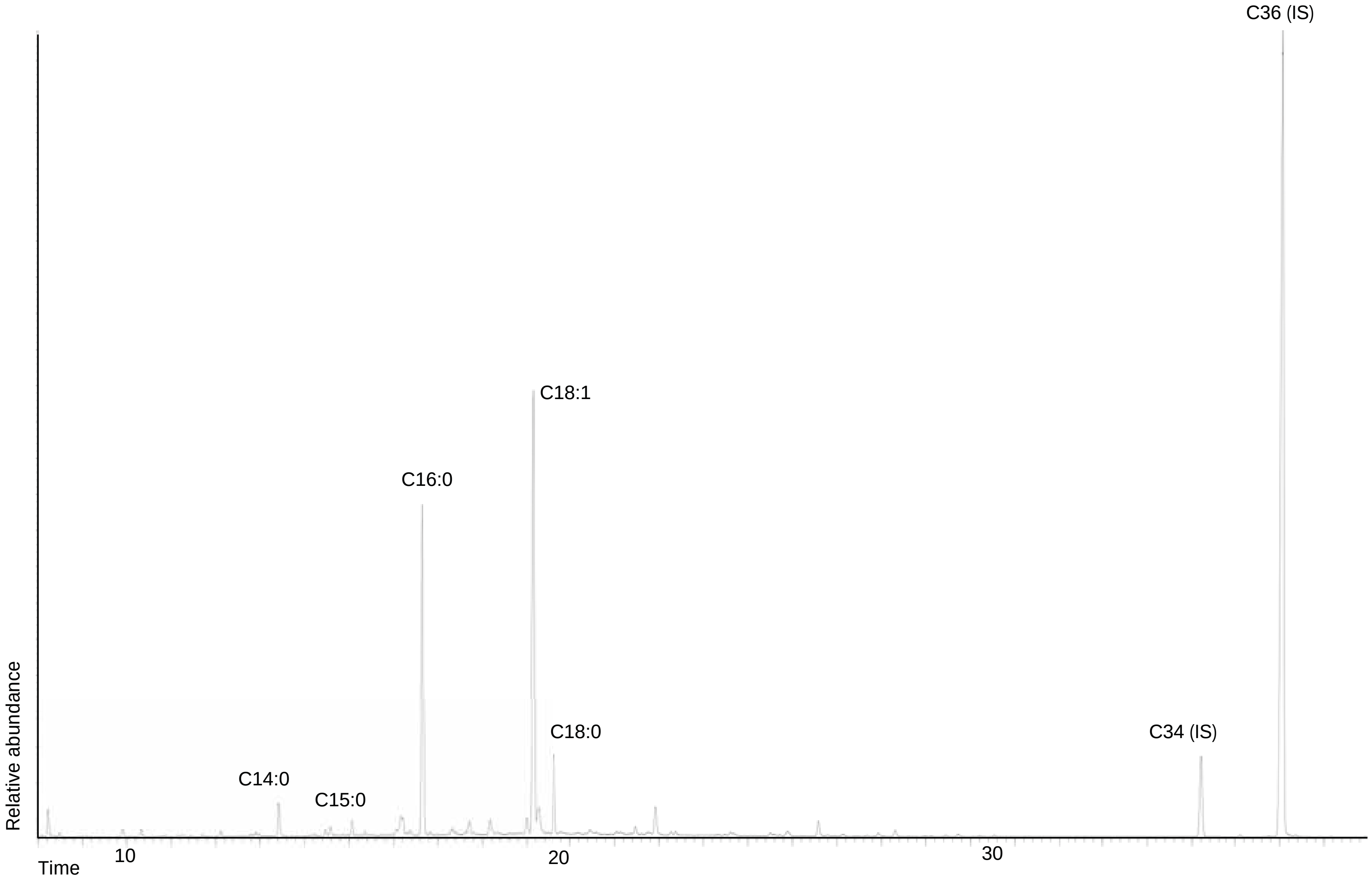
BF2


C36 (IS)
C16:0
C18:1
C18:0
C34 (IS)
C14:0
C15:0
Relative abundance
Time
10
20
30

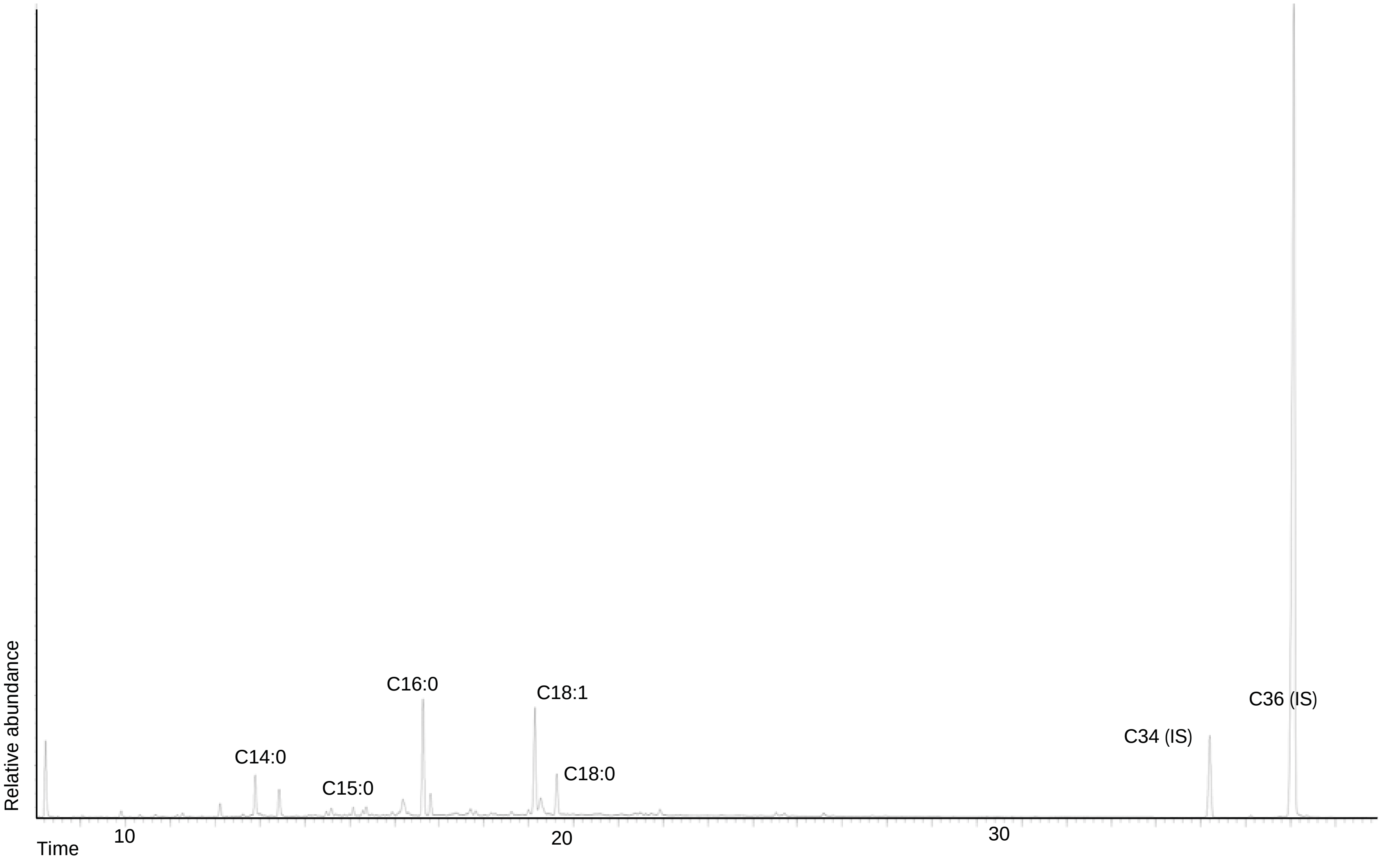
BF3


Relative abundance
C14:0
C15:0
C16:0
C18:1
C18:0
C34 (IS)
C36 (IS)
Time
10
20
30

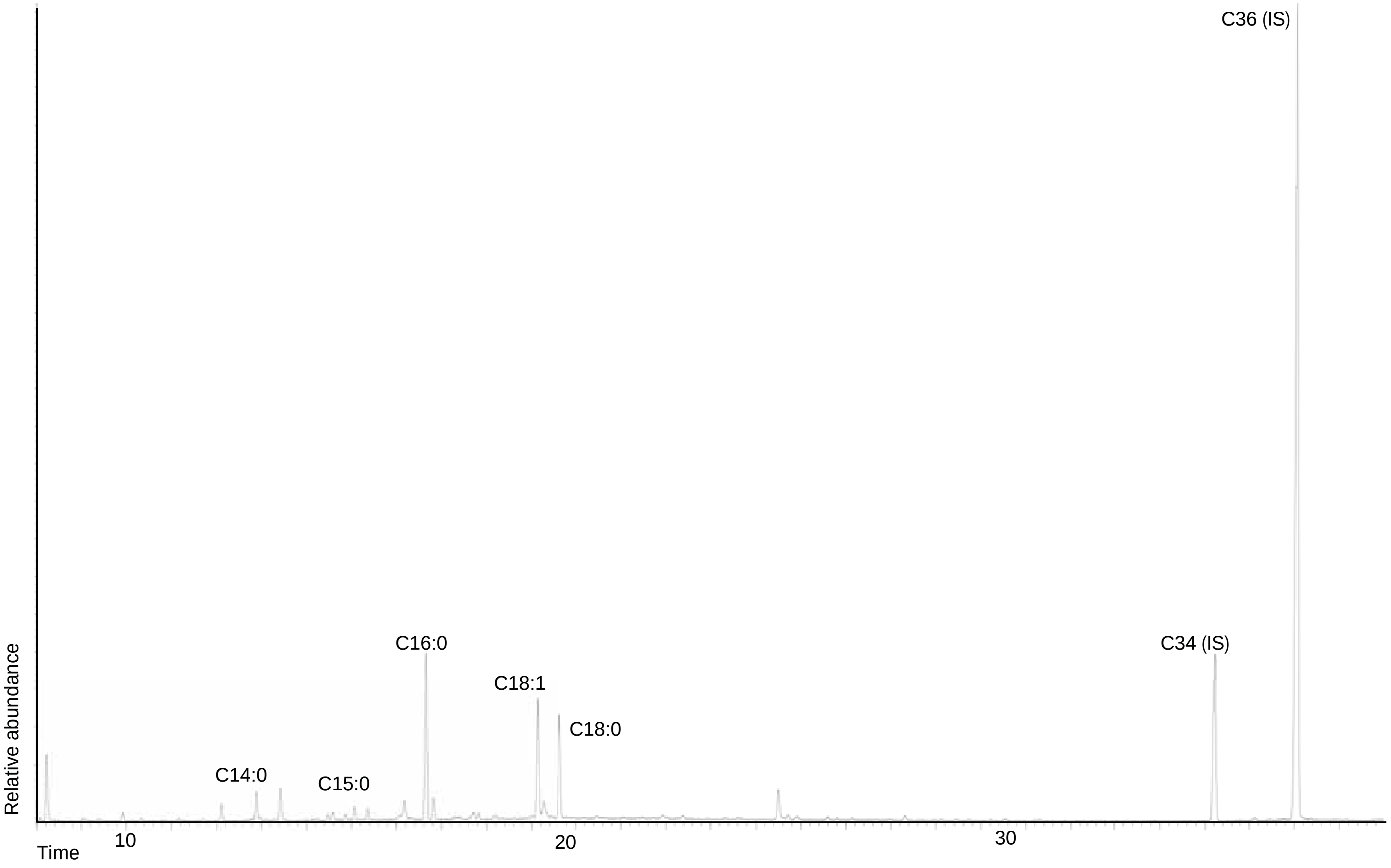
BF4


C36 (IS)
C18:1
C16:0
C18:0
C34 (IS)
C14:0
C15:0
Relative abundance
Time
10
20
30

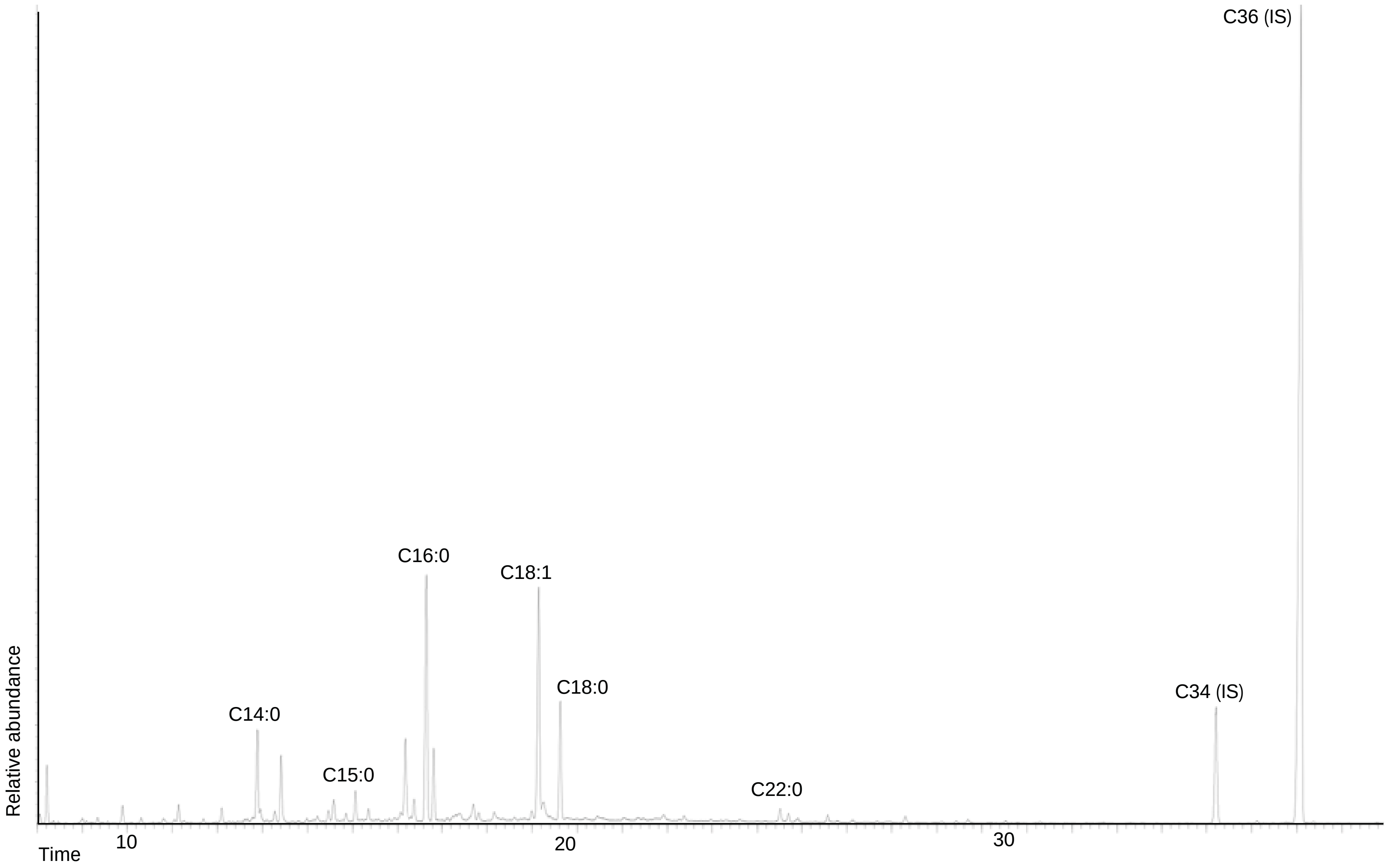
BF5


Relative abundance
C14:0
C15:0
C16:0
C18:1
C18:0
C34 (IS)
C36 (IS)
Time
10
20
30

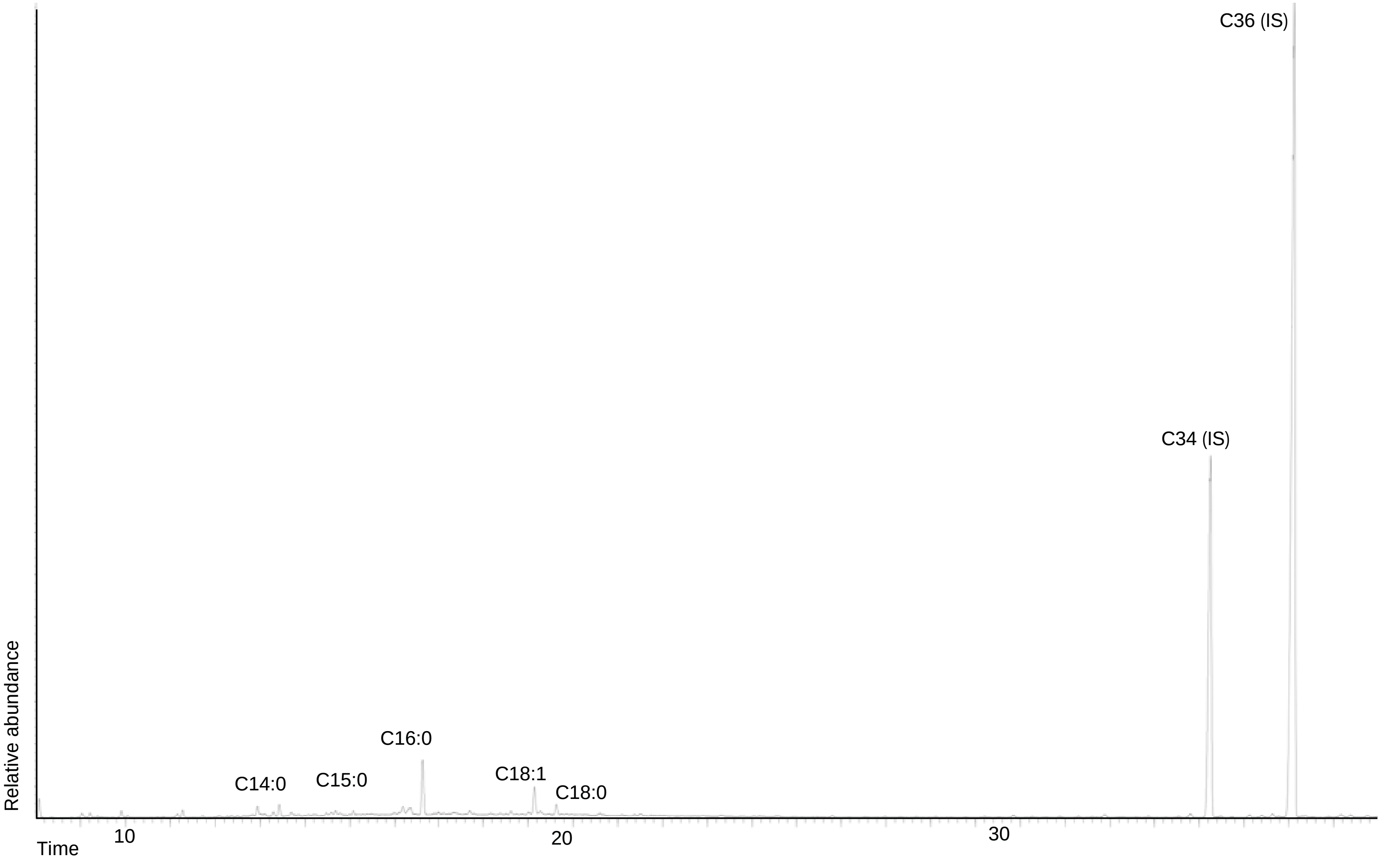
BF6


C36 (IS)
C34 (IS)
C16:0
C18:1
C18:0
C14:0
C15:0
Relative abundance
Time
10
20
30

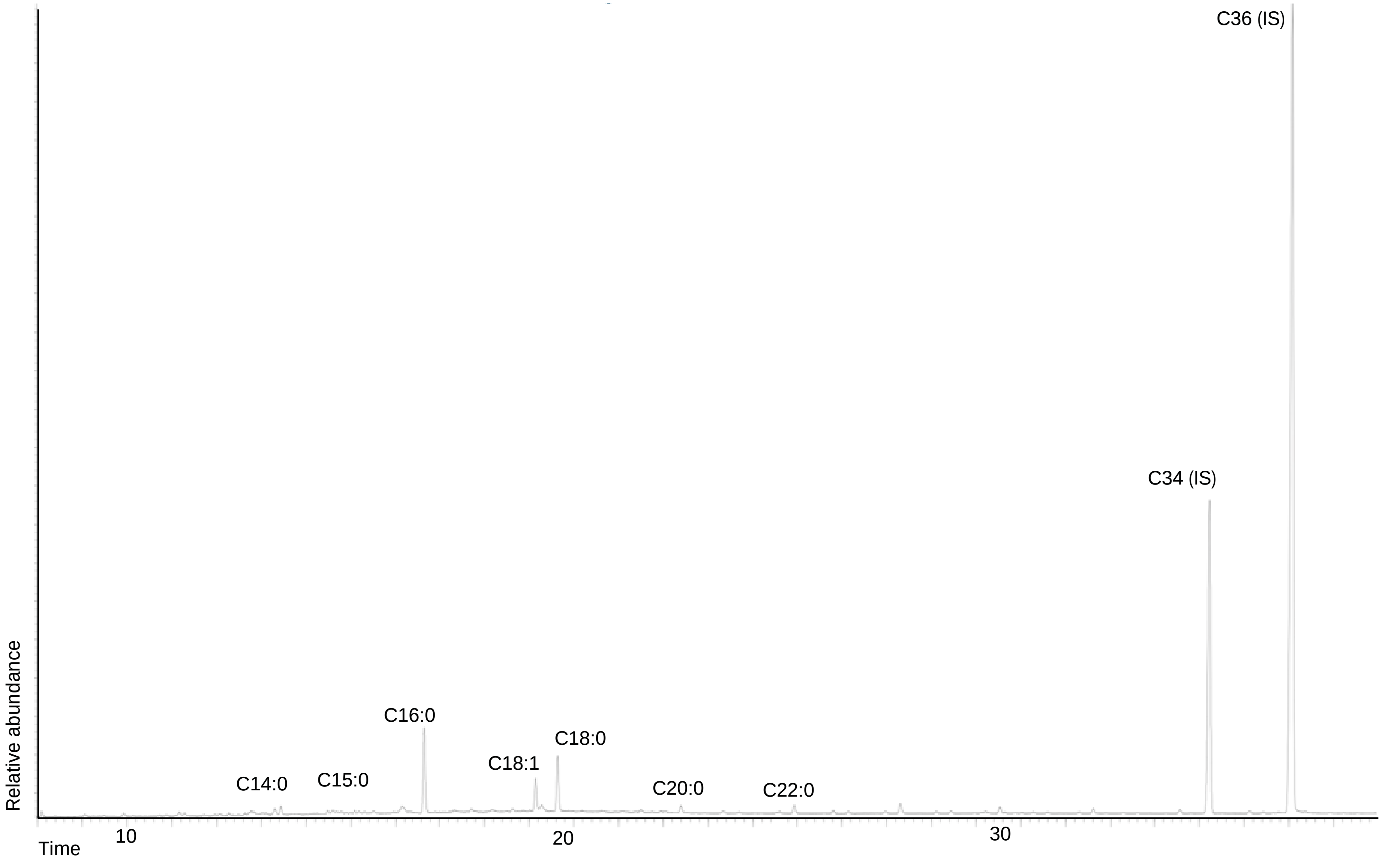
BF7


C36 (IS)
C16:0
C18:1
C18:0
C14:0
C34 (IS)
C15:0
C22:0
Relative abundance
Time
10
20
30

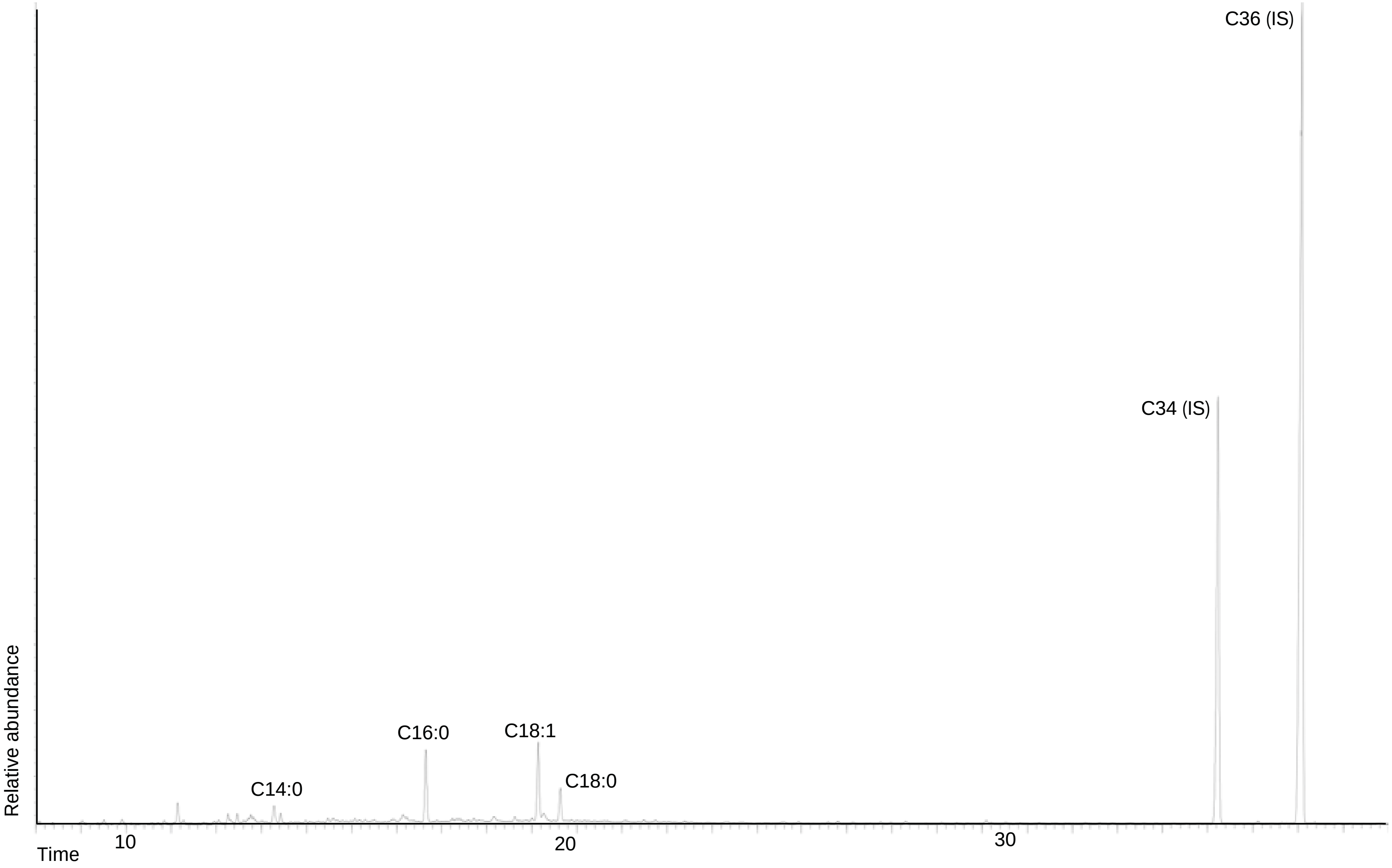
BF32


C36 (IS)
C34 (IS)
C16:0
C14:0
C15:0
C18:1
C18:0
Relative abundance
10
20
30
Time

BF45

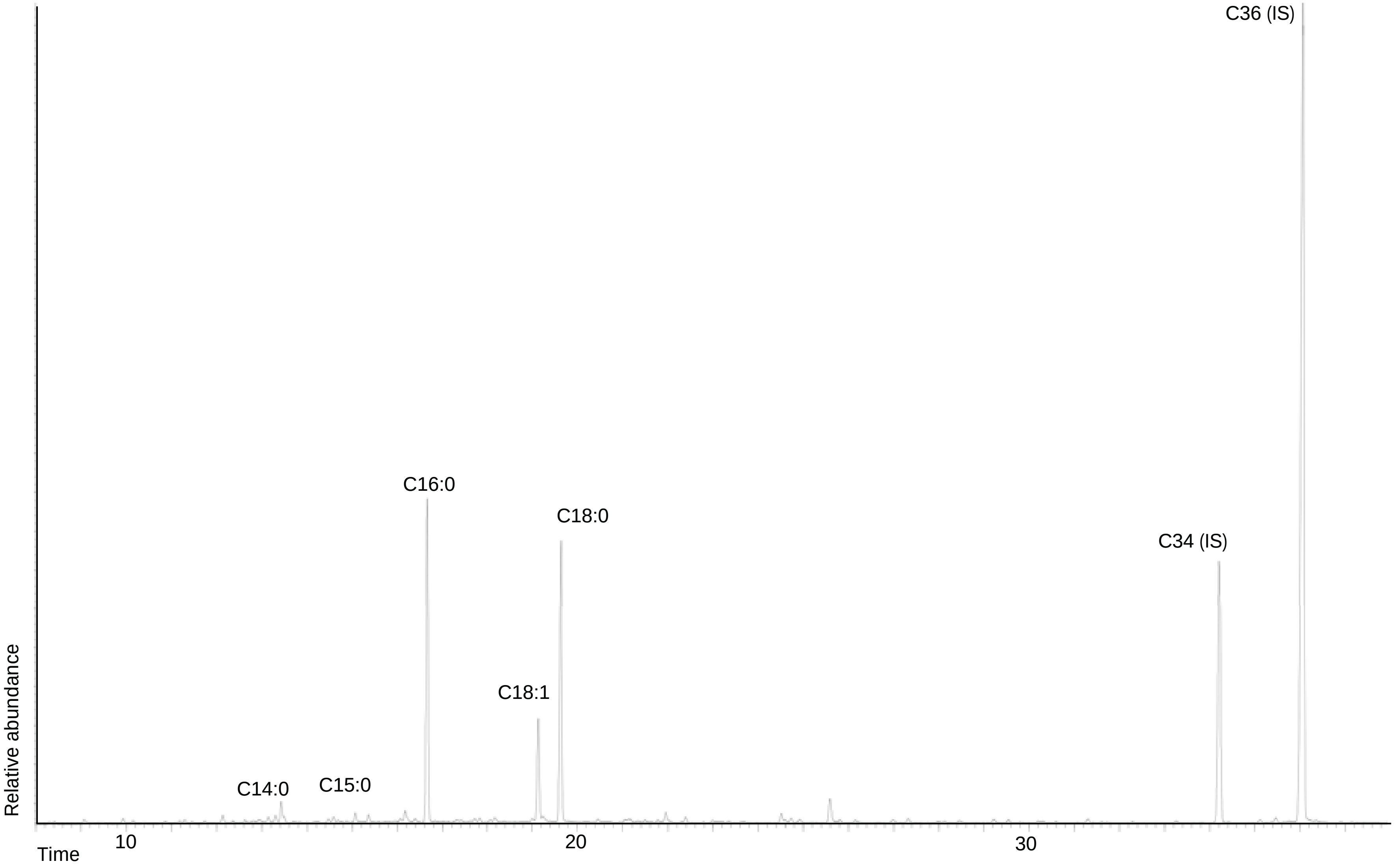

C36 (IS)
C34 (IS)
C16:0
C18:0
C18:1
C14:0
C15:0
C20:0
C22:0
Relative abundance
Time
10
20
30

BF46

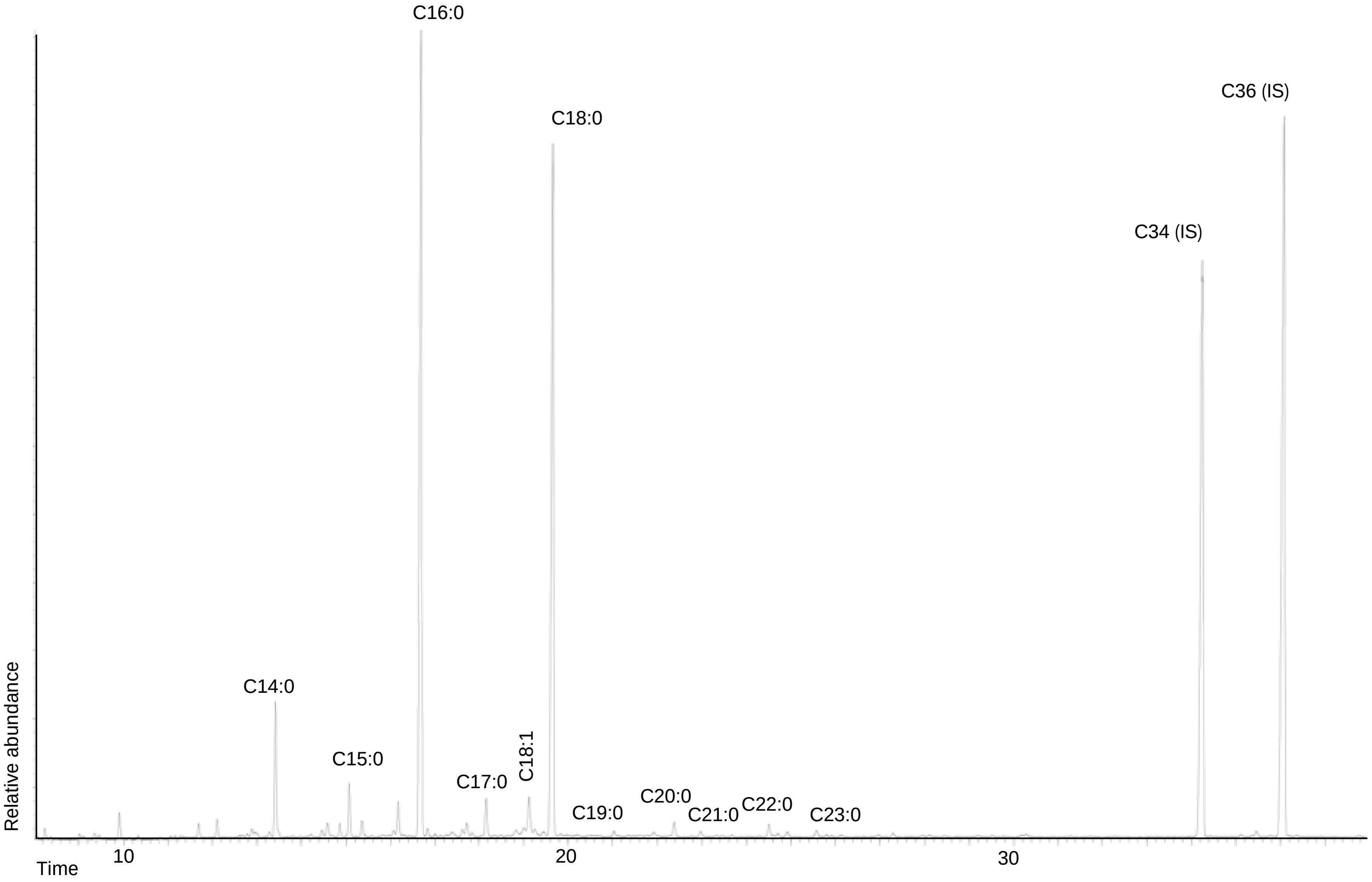

C36 (IS)
C34 (IS)
C16:0
C18:1
C18:0
C14:0
Relative abundance
Time
10
20
30

El Molló

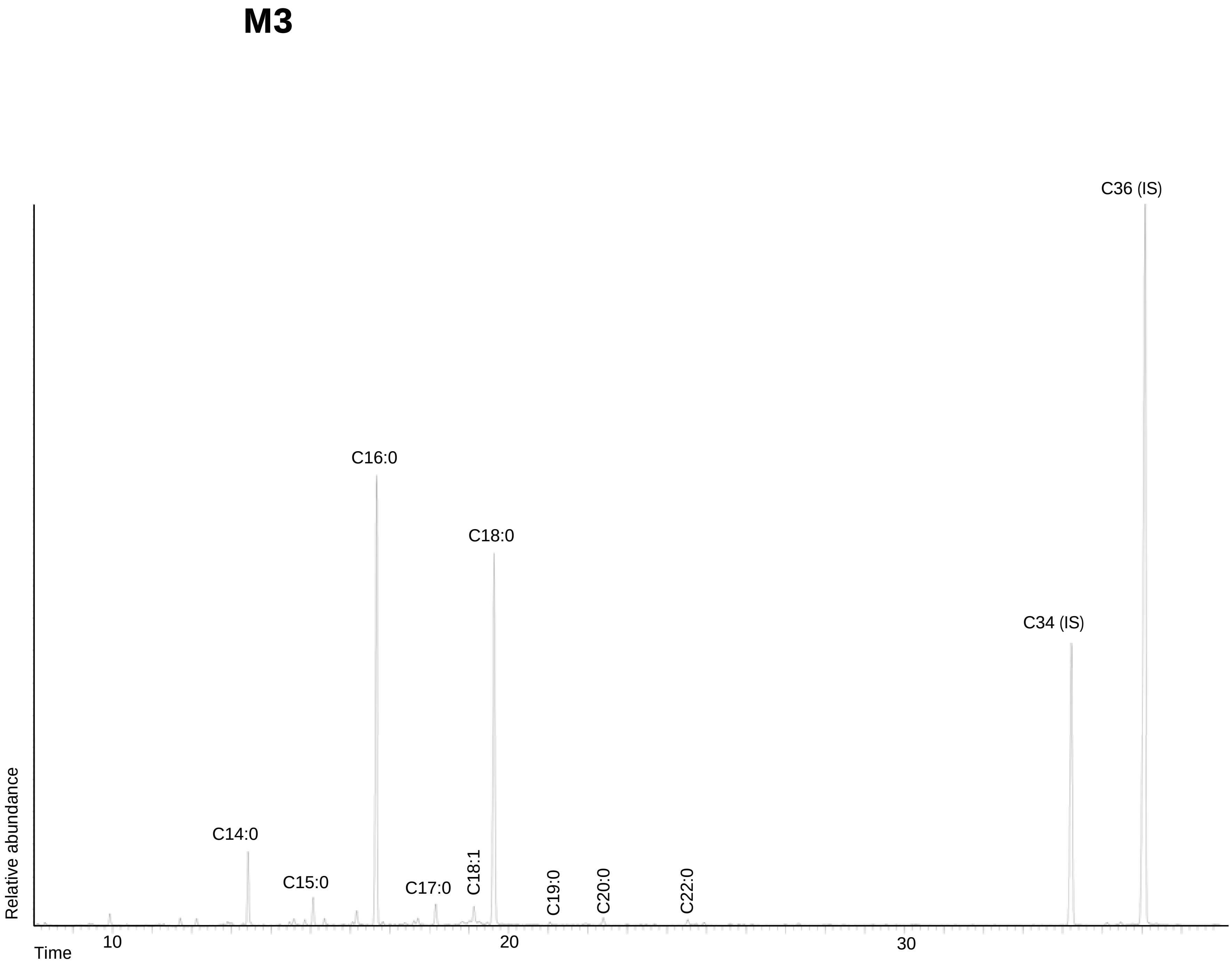
M1


C36 (IS)
C16:0
C18:0
C34 (IS)
C18:1
C14:0
C15:0
Relative abundance
Time
10
20
30

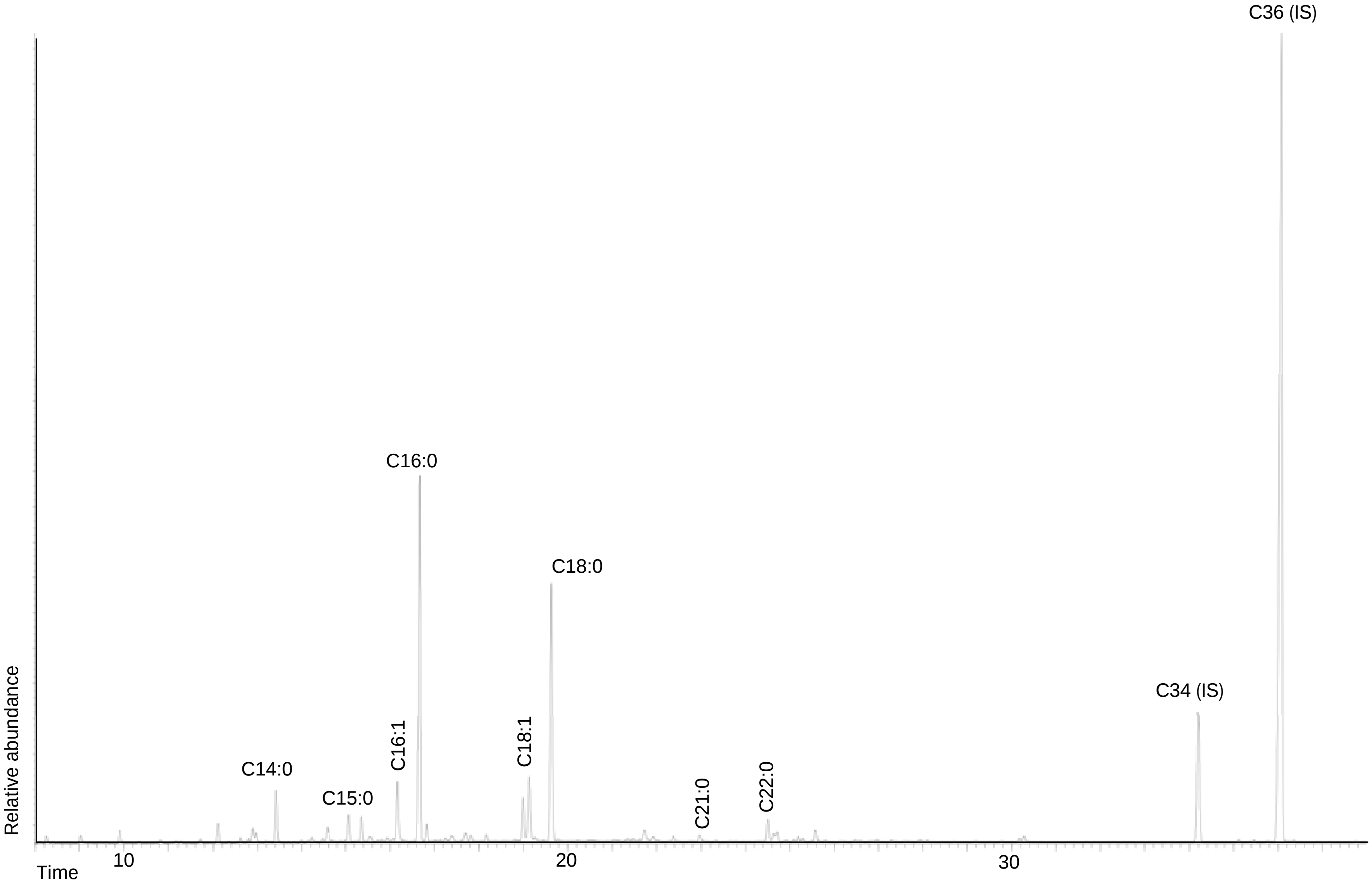
M2



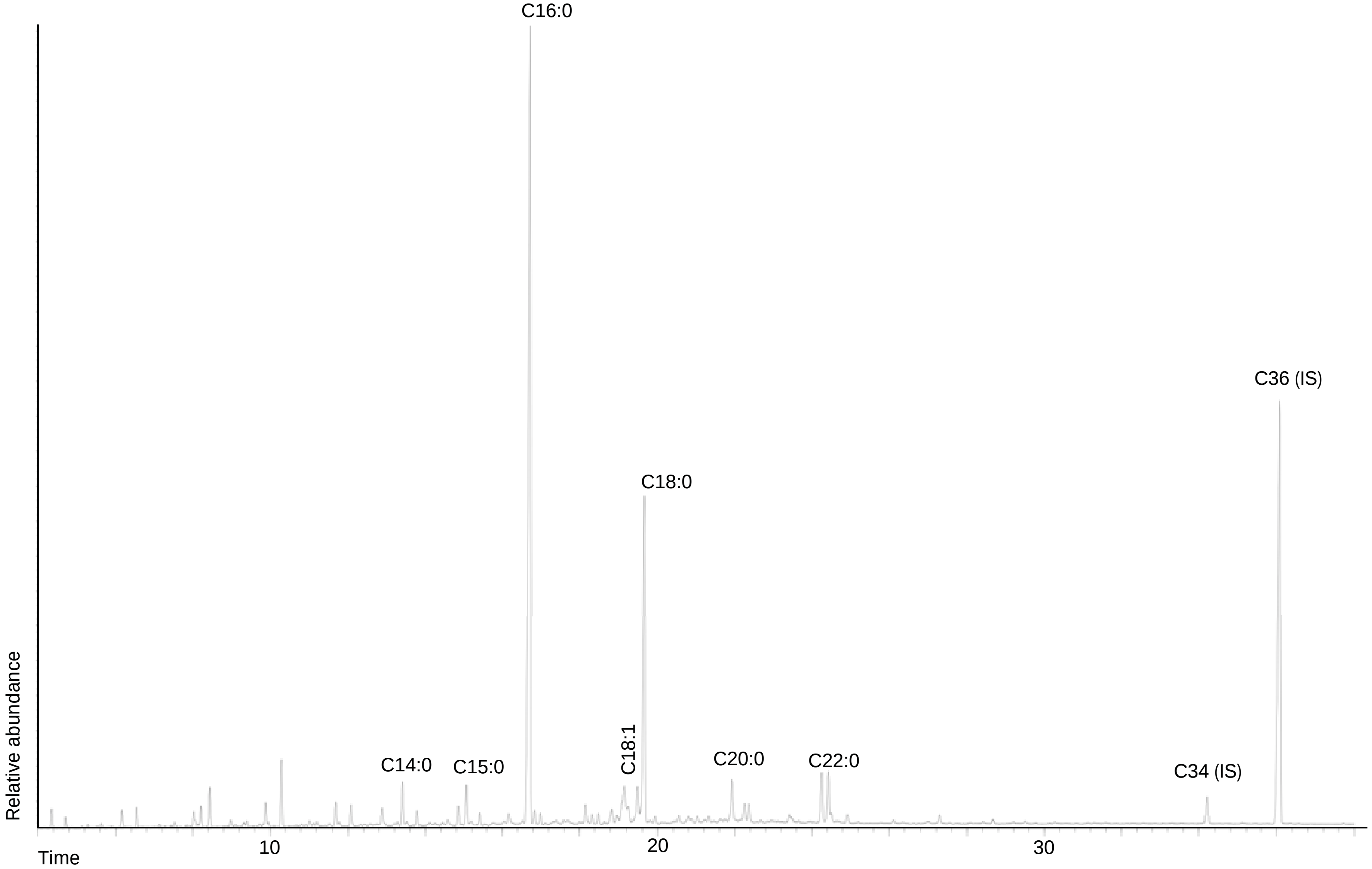
C16:0
C36 (IS)
C18:0
C34 (IS)
C14:0
C15:0
C17:0
C18:1
C19:0
C20:0
C21:0
C22:0
C23:0
Relative abundance
Time
10
20
30


M3
C36 (IS)
C16:0
C18:0
C34 (IS)
C14:0
C15:0
C17:0
C18:1
C19:0
C20:0
C22:0
Relative abundance
Time
10
20
30

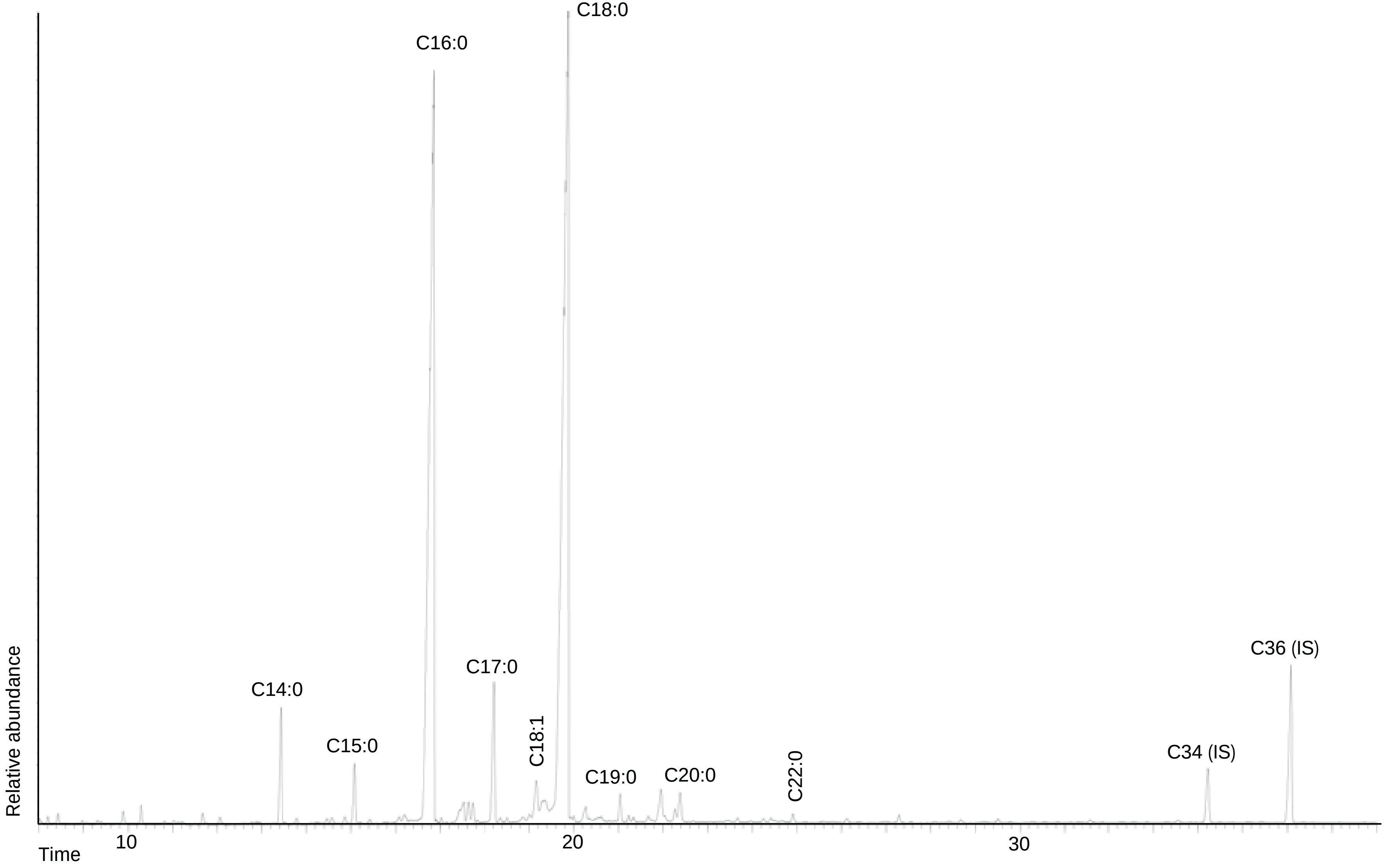
M4


C36 (IS)
C16:0
C18:0
C34 (IS)
C14:0
C15:0
C16:1
C18:1
C21:0
C22:0
Relative abundance
Time
10
20
30

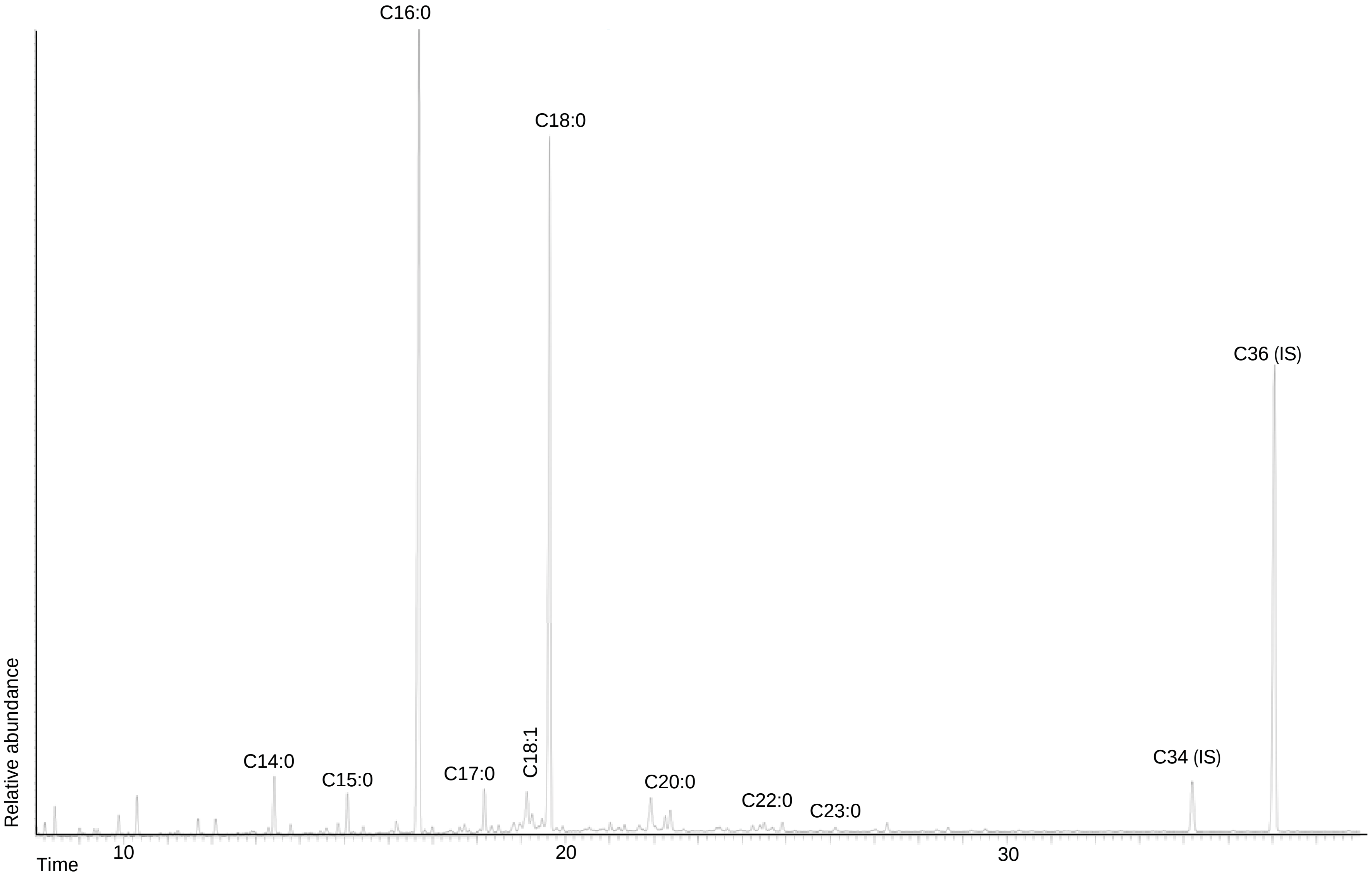
M5


C16:0
C36 (IS)
C18:0
C14:0
C15:0
C18:1
C20:0
C22:0
C34 (IS)
Relative abundance
Time
10
20
30

M6

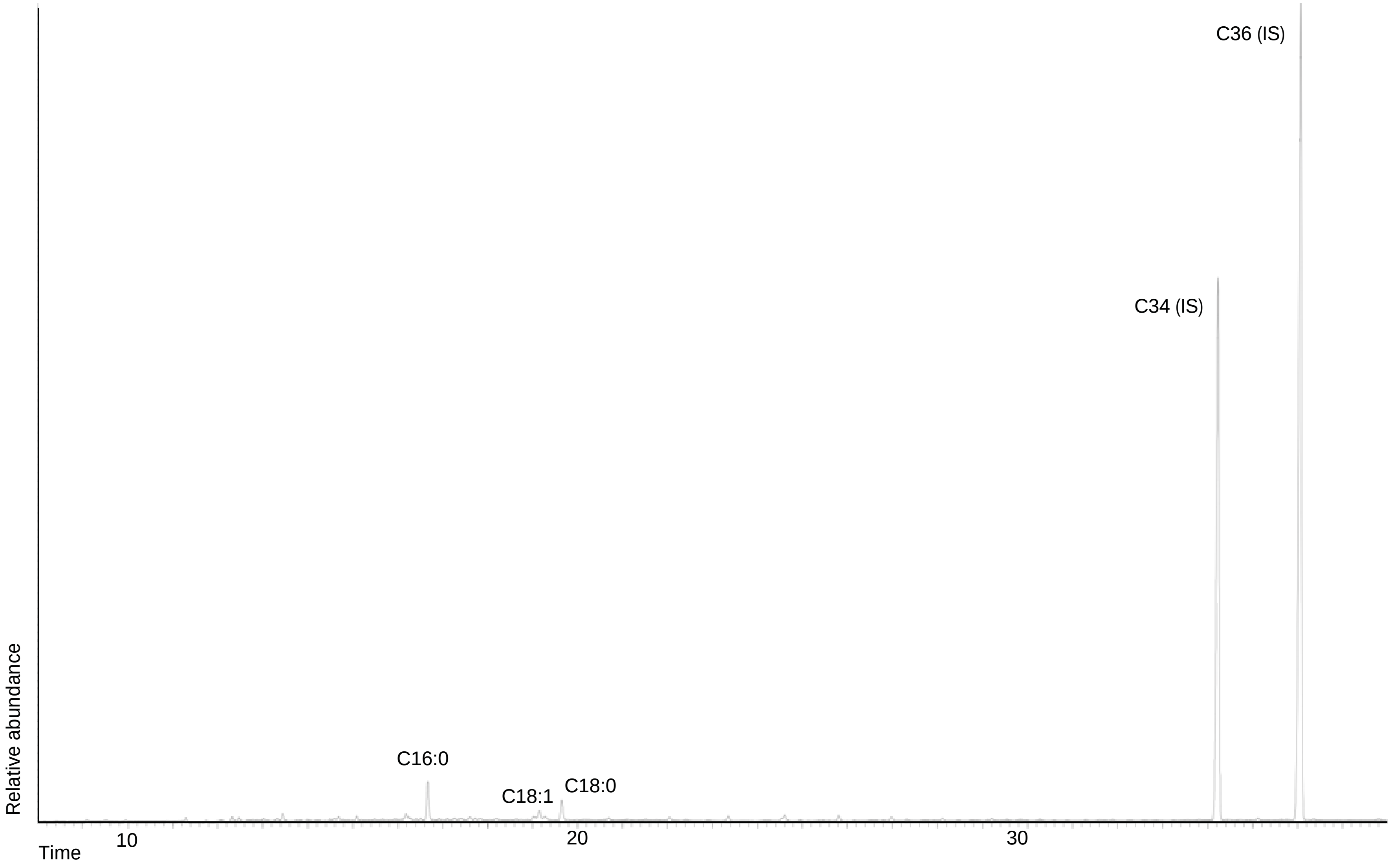

C18:0
C16:0
C17:0
C14:0
C15:0
C18:1
C19:0
C20:0
C22:0
C34 (IS)
C36 (IS)
Relative abundance
Time
10
20
30

M7

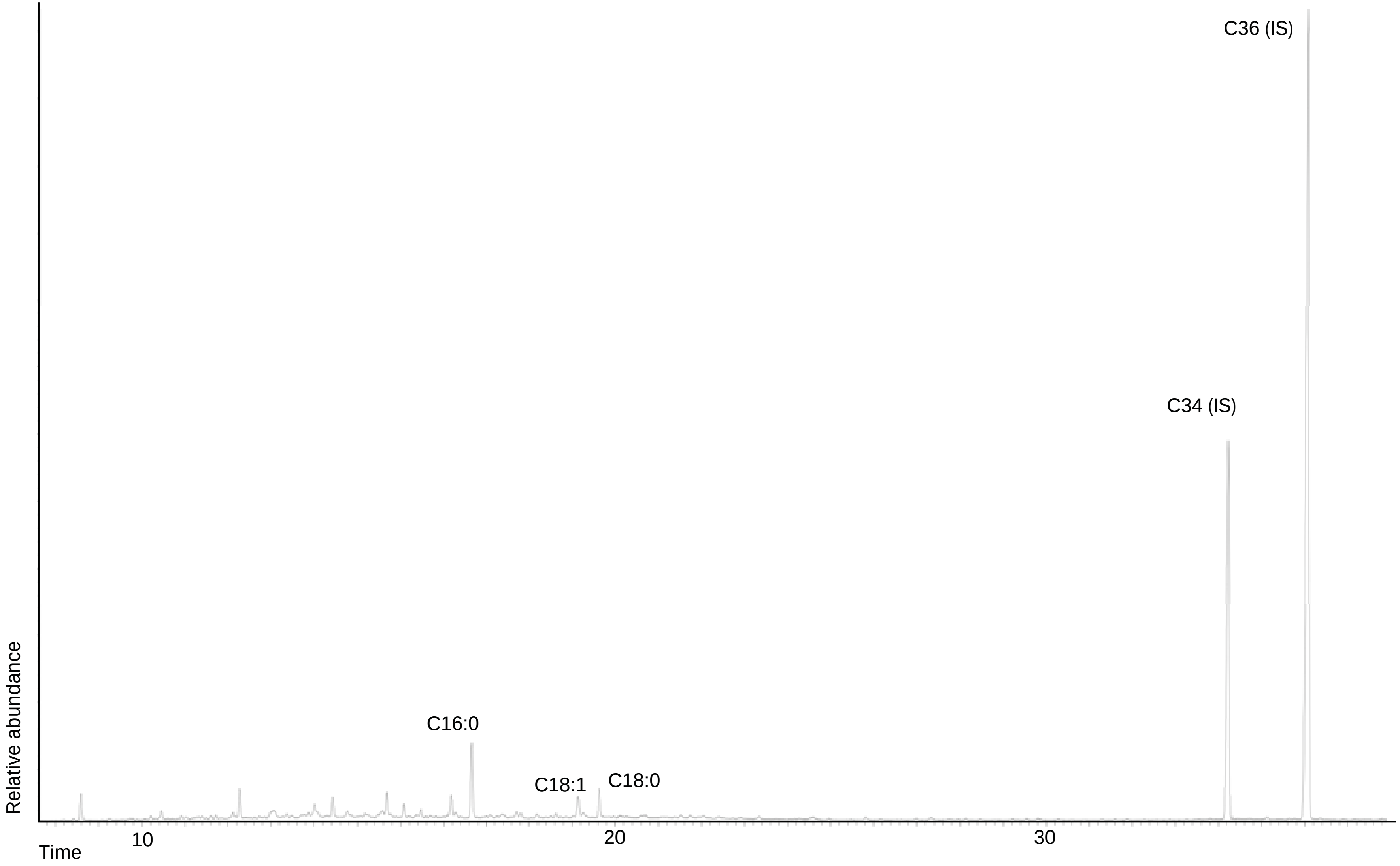

C16:0
C18:0
C36 (IS)
C34 (IS)
C14:0
C15:0
C17:0
C18:1
C20:0
C22:0
C23:0
Relative abundance
Time
10
20
30

El Cau de les Guilles

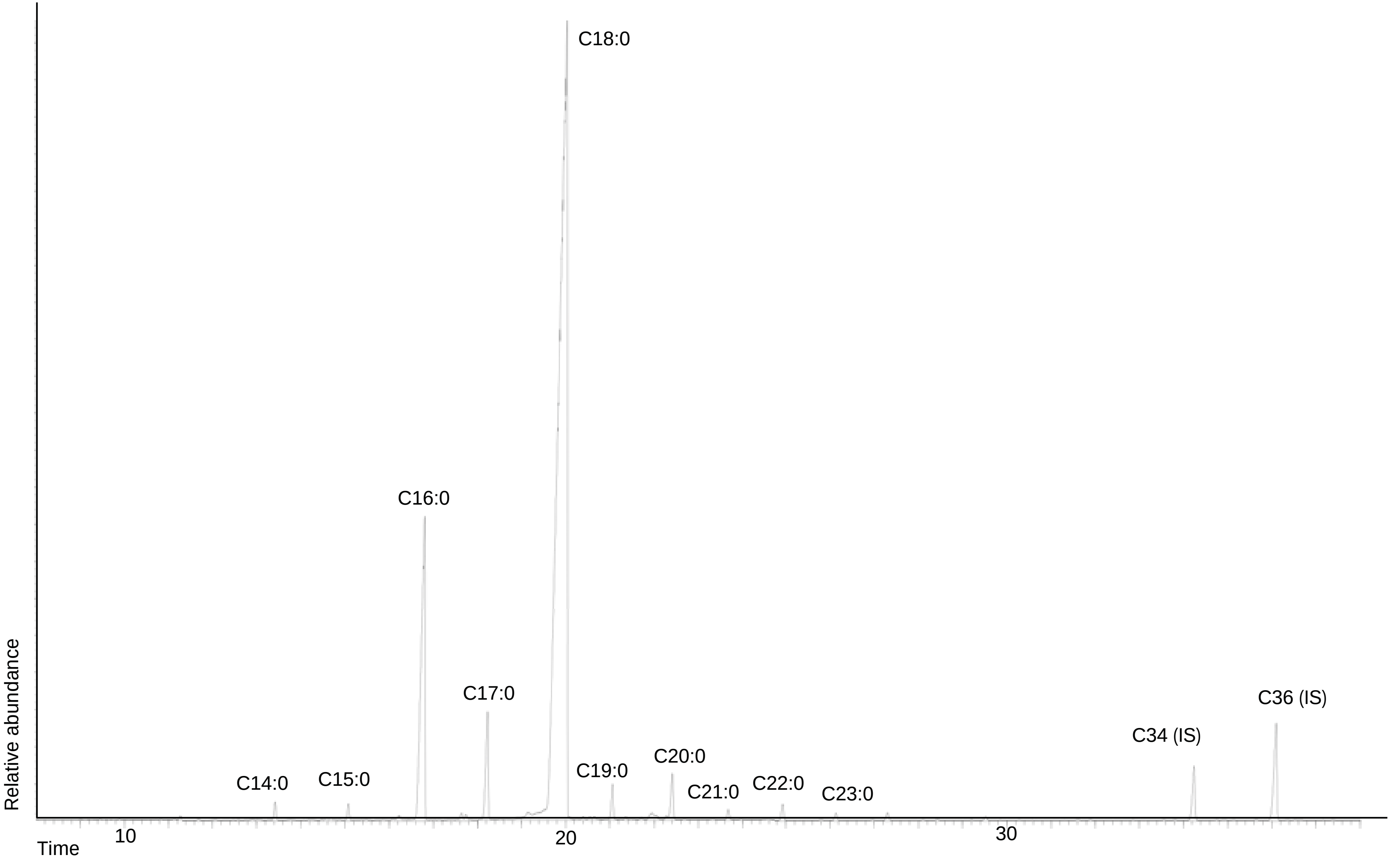
CGVI2 35



CGVI2 30



CGVI 23


C18:0
C16:0
C17:0
C14:0
C15:0
C19:0
C20:0
C21:0
C22:0
C23:0
C34 (IS)
C36 (IS)
Relative abundance
Time
10
20
30

CGVI 32