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SEDIMENT FINGERPRINTING AND HYDRO-SEDIMENTARY MONITORING AS TOOLS FOR CATCHMENT MANAGEMENT IN MEDITERRANEAN ENVIRONMENTS

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Doctor by the Universitat de les Illes Balears

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Preliminary results:

In-channel alterations of the most common soil properties used as tracers in sediment fingerprinting studies. **Paper in preparation**.

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List of acronyms

AR1d - One-day

AR3d - Three days antecedent rainfall

AR7d - Seven days antecedent rainfall

BNDVI - Blue Normalized Difference Vegetation Index

CV - Coefficient of variation

DEM - Digital Elevation Model

DFA - Discriminant Function Analysis

dNBR - differenced Normalized Burn Ratio

DOD - DEM of Difference

DS - Downstream Site

DTM - Digital Terrain Model

EMMA - End Member Mixing Models

FRNs - Fallout Radionuclides

GOF – Goodness of Fit

I30 – Rainfall intensity in 30 minutes

IPmax-30 - Maximum rainfall intensity in 30 min

LFF - Large Forest Fires

LIST - Luxembourg Institute of Science and Technology

Load - Total sediment load

nRMSE - normalized Root Mean Square Error

P.tot - Total rainfall

PCA - Principal Component Analysis

PSD - Particle Size Distribution

Q - Stream discharge

Qmax - Maximum discharge peak

Qmean - average discharge

Rtot – Total rainfall

SfM - Structure from Motion

SS - Suspended Sediment

SS peak - Maximum Sediment Concentration

SSA – Specific Surface Area

SSC - Suspended Sediment Concentrations

SSCmax - Maximum suspended sediment concentration

SSCmean - Mean suspended sediment concentration

SSload - Total suspended sediment load

US - Upstream Site

Wvol - Total water volume

Abstract

Soil erosion is a natural process that encompasses weathering, transport and deposition of soil particles. These processes are essential in terrestrial geochemical cycles. However, the on- and off-site erosion effects are considered to be one of the most important causes of terrestrial and aquatic ecosystems degradation. The characteristics of the Mediterranean region are marked by complex relationships between natural, human, biotic and abiotic variables. In addition, an irregular rainfall distribution, strong seasonality and the physiographic landscape characteristics promote divergent responses in erosion rates and sediment yields. In this context, the Mediterranean basin has the highest sediment yields in all of Europe. In addition, it is emerging as a hot spot point in Global Change dynamics, especially with reference to climate and land use change, which could generate an increase in erosive and sediment transport processes. At the catchment scale, sediment transfer occurs in hill slopes, between hill slopes and channels or within channels. Information on the nature and relative contribution of sediment sources is a key aspect with regard to designing and implementing erosion control strategies in catchments.

The main objective of this thesis is to identify erosion and sediment transport processes in two Mediterranean catchments affected by different global change processes at different spatio-temporal scales, improving current techniques for sediment origin determination (i.e., reducing uncertainties, time and cost) so it can better implemented in catchment management plans. For this purpose, the hydro-sedimentary dynamics and the origin of the sediments has been investigated on the island of Mallorca (Spain), in two small catchments; the Sa Font de la Vila catchment - 4.8 km^2, affected by wildfires - and the Es Fangar catchment (3.4 km^2), affected by land use changes. The combination of sediment fingerprinting and hydro-sedimentary monitoring made it possible to assess its hydro-sedimentary dynamics during the study period. In Sa Font de la Vila, results showed a gradual decrease in contribution from burned sources over time, while in Es Fangar the contributions from crops dominated throughout the study period, without substantial changes. Sediment yields were 6.3 t km^2 yr^{-1} and 4.5 t km^2 yr^{-1} for Sa Font de la Vila and Es Fangar respectively, low results in comparison with other Mediterranean catchments. This was mainly attributed to the calcareous lithology, land uses (in Es Fangar catchment), vegetation recovery (in Sa Font de la Vila catchment) and agricultural terraces.

The use of soil colour parameters as tracers was successfully evaluated in the two catchments, confirming its suitability as a fast and inexpensive tracer, even in fire-affected catchments. Furthermore, the strong correlations between the measurements made with a spectro-radiometer and a scanner make colour even more accessible for its implementation in catchment management plans. The experiment on tracer conservatism confirmed that in-channel changes suffered by all the analysed tracers (coefficient of variation $\bar{x}$ 8.1 ± 8.8%) were generally lower than their spatial variability within the catchment (coefficient of variation $\bar{x}$ 16.3 ± 18.5%). Furthermore, the colour parameters were the least variable tracers (i.e. the most conservative). with a coefficient of variation of 2.6 ± 2.2%.

Finally, it was not possible to identify the activation patterns of different sediment sources combining hydro-sedimentary monitoring and sediment fingerprinting. This was probably caused by Es Fangar's catchment stability in terms of the origin of the suspended sediment. Es Fangar catchment sediment source stability is attributed to lithological characteristics, land uses and the presence of agricultural terraces in the study area. However, events of higher magnitude could exceed the sedimentary (dis)connectivity thresholds of the rest of the sources, promoting a sediment cascade effect.

The results presented in this thesis are relevant and represent an advance in the optimization of the sediment fingerprinting technique. Despite some limitations that need to be further investigated, hydro-sedimentary monitoring and sediment fingerprinting used in combination was shown to be very useful for integrated catchment management plans in Mediterranean environments.

Key words: Sediment fingerprinting, Mediterranean catchments, hydro-sedimentary monitoring, catchment management.

Resumen

La erosión es un proceso natural que comprende la meteorización, transporte y depósito de partículas del suelo. Estos procesos son esenciales dentro de los ciclos geoquímicos terrestres. Sin embargo, los efectos *in situ* y *ex situ* de la erosión se consideran una de las causas más importantes de la degradación de la calidad en ecosistemas terrestres y acuáticos. Las características de la región mediterránea están marcadas por relaciones complejas entre variables naturales, humanas, bióticas y abióticas. Además, una distribución irregular de las lluvias, una marcada estacionalidad y las características fisiográficas del paisaje promueven respuestas divergentes en las tasas de erosión y producción de sedimentos. En este contexto, la cuenca Mediterránea presenta los rendimientos de sedimento más altos de toda Europa. Además, se perfila como un punto crítico de la dinámica del Cambio Global, especialmente en lo que respecta al Cambio Climático y de uso del suelo, lo que podría generar un aumento de los procesos erosivos y de transporte de sedimento. A escala de cuenca de drenaje, la transferencia de sedimentos ocurre en laderas, entre laderas y canales o dentro de canales. La información sobre la naturaleza y contribución relativa de las fuentes de sedimento es un aspecto clave para diseñar e implementar estrategias de control de la erosión en cuencas de drenaje.

El objetivo principal de esta tesis es identificar procesos de erosión y transporte de sedimentos en dos cuencas mediterráneas afectadas por diferentes procesos de cambio global a diferentes escalas espacio-temporales, mejorando las técnicas actuales para la determinación del origen de los sedimentos (es decir, reducir incertidumbres, tiempo y costo) para su mejor implementación en planes de gestión de cuencas de drenaje. Para ello, se investigó la dinámica hidro-sedimentaria y el origen de los sedimentos en dos pequeñas cuencas de drenaje de la isla de Mallorca (España); Sa Font de la Vila -4,8 km^2, afectada por incendios forestales - y Es Fangar (3,4 km^2), afectada por cambios de usos del suelo. La combinación de la técnica sediment fingerprinting y monitoreo hidro-sedimentario continuo permitió evaluar su dinámica hidro-sedimentaria durante el período de estudio. En Sa Font de la Vila, los resultados mostraron una disminución paulatina de las aportaciones de fuentes quemadas a lo largo del tiempo, mientras que en Es Fangar las aportaciones de las zonas de cultivos dominaron durante todo el período de estudio sin cambios sustanciales. Los rendimientos de sedimentos fueron 6,3 t km^2 a^{-1} y 4,5 t km^2 a^{-1} para Sa Font de la Vila y Es Fangar respectivamente, bajos en comparación con otras cuencas mediterráneas. Esto se atribuyó principalmente a la litología calcárea de las cuencas, los usos del suelo (en Es Fangar), la recuperación de la vegetación (en Sa Font de la Vila) y la presencia de terrazas agrícolas.

El uso de parámetros de color como trazadores se evaluó con éxito en las dos cuencas, lo que confirma su idoneidad para su uso como un trazador rápido y económico, incluso en cuencas afectadas por incendios. Además, las fuertes correlaciones entre las medidas tomadas con un espectro-radiómetro y un escáner, hacen del color un trazador muy accesible para su implementación en planes gestión. El experimento sobre conservación de las propiedades de los trazadores mostró variaciones bajas en la mayoría de los trazadores analizados (coeficiente de variación $\bar{x}$ 8,1 ± 8,8%). Estas fueron generalmente menores que su propia variabilidad espacial dentro de la cuenca (coeficiente de variación x̄ 16,3 ± 18,5%). Además, los parámetros de color fueron los trazadores menos variables (i.e. más conservadores) con un coeficiente de variación de 2,6 ± 2,2%.

Finalmente, no fue posible identificar los patrones de activación de diferentes fuentes de sedimentos combinando el monitoreo hidro-sedimentario y sediment fingerprinting. Esto fue causado principalmente por la estabilidad de la cuenca de Es Fangar en términos de origen de sedimentos en suspensión. La estabilidad de las fuentes de sedimentos se atribuyó a las características litológicas, usos del suelo y la presencia de terrazas agrícolas en el área de estudio. Sin embargo, eventos de mayor magnitud podrían superar los umbrales de (des)conectividad sedimentaria del resto de fuentes consideradas y activarlas.

Los resultados que presenta esta tesis son relevantes y suponen un avance en la optimización de la técnica sediment fingerprinting. Pese a algunas limitaciones que se han de seguir investigando, se demostró que la combinación de monitoreo hidro-sedimentario y sediment fingerprinting es de gran utilidad para los planes de gestión integrada de cuencas de drenaje Mediterráneas.

Palabras clave: Sediment fingerprinting, Cuencas mediterráneas, monitoreo hidro-sedimentario, Gestión de cuencas de drenaje.

Resum

L'erosió és un procés natural que comprèn la meteorització, transport i dipòsit de partícules sòl. Aquests processos són essencials dins dels cicles geoquímics terrestres. No obstant això, els efectes *in situ* i *ex situ* de l'erosió es consideren una de les causes més importants de la degradació de la qualitat en ecosistemes terrestres i aquàtics. Les característiques de la regió mediterrània estan marcades per relacions complexes entre variables naturals, humanes, biòtiques i abiòtiques. A més, una distribució irregular de les pluges, una marcada estacionalitat i les característiques fisiogràfiques del paisatge promouen respostes divergents en les taxes d'erosió i la producció de sediments. En aquest context, la conca Mediterrània presenta els rendiments de sediment més alts de tot Europa. A més, es perfila com un punt crític de la dinàmica del Canvi Global, especialment pel que fa a el Canvi Climàtic i usos del sòl, la qual cosa podria generar un augment dels processos erosius i de transport de sediment. A escala de conca de drenatge, la transferència de sediments es dona en vessants, entre vessants i canals o dins els canals. La informació sobre la naturalesa i contribució relativa de les fonts de sediment és un aspecte clau per dissenyar i implementar estratègies de control de l'erosió en conques de drenatge.

L'objectiu principal d'aquesta tesi és identificar processos d'erosió i transport de sediments en dues conques mediterrànies afectades per diferents processos de Canvi Global a diferents escales espai-temporals, millorant les tècniques actuals per a la determinació de l'origen dels sediments (és a dir, reduir incerteses, temps i cost) per a la seva millor implementació en plans de gestió de conques de drenatge. Per a això, es va investigar la dinàmica hidro-sedimentària i l'origen dels sediments en dos petites conques de drenatge de l'illa de Mallorca (Espanya); Sa Font de la Vila -4,8 km^2, afectada per incendis forestals - i Es Fangar (3,4 km^2), afectada per canvis d'usos del sòl. La combinació de la tècnica sediment fingerprinting i monitoratge hidro-sedimentari va permetre avaluar la seva dinàmica hidro-sedimentària durant el període d'estudi. A Sa Font de la Vila, els resultats van mostrar una disminució gradual de les aportacions de fonts cremades al llarg del temps, mentre que a Es Fangar les aportacions de fonts de cultius van dominar durant tot el període d'estudi sense canvis substancials. Els rendiments de sediments van ser 6,3 t km^2 a^{-1} i 4,5 t km^2 a^{-1} per a Sa Font de la Vila i Es Fangar respectivament, baixos en comparació amb altres conques mediterrànies. Això es va atribuir principalment a la litologia calcària de les conques hidrogràfiques, els usos de terra (a Es Fangar), la recuperació de la vegetació (a Sa Font de la Vila) i la presència de terrasses agrícoles.

L'ús de paràmetres de color com a traçadors es va avaluar amb èxit en les dues conques, la qual cosa confirma la seva idoneïtat per al seu ús com un traçador ràpid i econòmic, fins i tot en conques afectades per incendis. A més, les fortes correlacions entre les mesures preses amb un espectre-radiòmetre i un escàner, fan del color un traçador molt accessible per a la seva implementació en plans gestió. L'experiment sobre conservació de les propietats dels traçadors, va mostrar variacions baixes en la majoria dels traçadors analitzats (coeficient de variació $\bar{x}$ 8,1 ± 8,8%). Aquestes van ser generalment menors que la seva pròpia variabilitat espacial dins de la conca (coeficient de variació $\bar{x}$ 16,3 ± 18,5%). A més, els paràmetres de color van ser els traçadors menys variables (i.e. més conservadors) amb un coeficient de variació de 2,6 ± 2,2%.

Finalment, no va ser possible identificar els patrons d'activació de diferents fonts de sediments combinant el monitoratge hidro-sedimentari i sediment fingerprinting. Això va ser causa principalment a l'estabilitat de la conca d'Es Fangar en termes d'origen de sediments en suspensió. L'estabilitat de les fonts de sediments s'atribueix a les característiques litològiques, usos de sòl i la presència de terrasses agrícoles en l'àrea d'estudi. No obstant això, esdeveniments de major magnitud podrien superar els llindars de (des)connectivitat sedimentària de la resta de fonts considerades i activar-les.

Els resultats que presenta aquesta tesi són rellevants i suposen un avanç en l'optimització de la tècnica sediment fingerprinting. Malgrat algunes limitacions que s'han de seguir investigant, es va demostrar que la combinació de monitorització hidro-sedimentari i sediment fingerprinting és de gran utilitat per als plans de gestió integrada de conques de drenatge Mediterrànies.

Paraules clau: Sediment fingerprinting, conques mediterrànies, monitorització hidro-sedimentària, gestió de conques de drenatge.

1. Introduction

1.1. Soil erosion and sediment transport processes in drainage catchments

Soil erosion is a natural process that encompasses the detachment, transport and deposition of soil particles driven by a specific force (i.e. water, wind, etc.). Thus, soil erosion plays a key role in the geological cycle (e.g. Garrels and Mackenzie, 1971; Wold and Hay, 1990), terrestrial geochemical cycles (e.g. Berhe et al., 2007; López-Bermúdez, 1990; Ludwig and Probst, 1996), aquatic ecosystems (e.g. Kjelland et al., 2015; Newcombe and Macdonald, 1991) as well as coastal areas and delta evolution (e.g. McLaughlin et al., 2003). However, erosion on-site effects are considered to be one of the most significant causes of soil quality degradation in natural, agricultural, and forest ecosystems and, therefore, in crop yield reduction (Pimentel and Kounang, 1998). Erosion global predictions, based in high spatial resolution Revised Universal Soil Loss Equation (RUSLE)-based semi-empirical modelling approach (GloSEM), determined global erosion in potential soil erosion rates of $43^{+9.2}_{-7}$ Pg yr^{-1} for 2015 (Borrelli et al., 2020). In addition, climatic predictions indicate an evolution of the hydrological cycle that can promote an increase of global water erosion processes around 30 to 66% (Borrelli et al., 2020). These trends in terrestrial systems, combined with an accelerated population growth, point erosion as a serious worldwide environmental and a human health issue (Pimentel, 2006).

The term Catchment is used in British English as a synonym for a river basin, whereas watershed is more associated with the line dividing two river basins. Therefore, a catchment is a topographical unit delimited by drainage divide watershed that isolates a stream system. All the surface area of a catchment drains to the same point, and it works as a "*hydrological response unit, a biophysical unit, and a holistic ecosystem in terms of the materials, energy, and information that flow through it*" (Wang et al., 2016). Drainage catchments integrate all aspects of the hydrological cycle as well as erosion and sediment transport processes from sources to sinks within a defined area (Sivapalan, 2005). Therefore, catchments are the fundamental landscape unit when it comes to the study of the cycling of water, sediments, and dissolved geochemical and biogeochemical constituents.

The sediment connectivity concept was defined by Bracken et al. (2015) as "*the connected transfer of sediment from a source to a sink in a system via sediment detachment and sediment transport, which is controlled by how the sediment moves between all geomorphic zones: on hill slopes, between hill slopes and channels and within channels*". Sediment connectivity is ruled by catchment structural features (i.e. structural connectivity), determined by its physical characteristics (e.g. morphology) and functional features, related with how the structural features interact through runoff processes (e.g. runoff generation, sediment transference between catchment compartments; i.e. functional connectivity) (Bracken et al., 2015; Najafi et al., 2021; Turnbull et al., 2008; Wainwright et al., 2011). Structural connectivity can be assessed by applying contiguity indexes, as indices of connectivity (e.g. Borselli et al., 2008; Cavalli et al., 2013; Heckmann et al., 2018). However, functional connectivity (or process-based connectivity; Bracken et al., 2015) is generally more difficult to measure (Calsamiglia et al., 2020; Wainwright et al., 2011). The complexity of the latter lies in the fact that it is dependent on the characteristics of the processes that connect the different structural catchment units, and therefore, on the magnitude of the events and their spatio-temporal distribution. An effective hillslope-to-channel connectivity generates a transference of eroded soil particles to the river network, introducing sediment into the river system. Size, mass and shape of the sediment particles, in combination with flow characteristics determine its transport. Within the channel system, coarse sediment particles (i.e. boulders to sand fraction) are transported in the lower part of the water column through rolling or saltation mechanisms, whereas, finer fractions (i.e. clay and silt) and dissolved sediment are transported in suspension within the turbulent flow (Hjulström, 1936). Fluxes altered by fine sediment particles transported downstream might modify physical, chemical and biotic processes in water bodies as, for example, altering light penetration and temperature, inducing siltation processes or increasing concentrations of nutrient, heavy metals or pesticides (Bilotta and Brazier, 2008; Collins and Walling, 2007). Similarly, altered fluxes can negatively affect aquatic ecosystems status (Newcombe and Macdonald, 1991; Verkaik et al., 2013), water quality (Horowitz et al., 2007), induce dam siltation and reduce the capacity of water reservoirs (Navas et al., 2004; Vörösmarty et al., 2003).

It is estimated that 95% of the sediment reaching oceans is transported by rivers (Syvitski, 2003) and suspended sediment equals 70% of the of the total sediment load (Morgan, 2005). Suspended sediment in many rivers normally encompass <2 mm fraction, with most of the load being <63 µm (Droppo, 2001; Phillips and Walling, 1995; Walling et al., 2000; Walling and Moorehead, 1989). The latest fraction (i.e. <63 µm) is considered the most chemically active component of the whole solid load (Foster and Charlesworth, 1996; Horowitz et al., 1993). In addition, suspended sediment is mainly transported in aggregate/flocculate form (Droppo et al., 1997; Droppo and Ongley, 1994). Flocs represent a complex interaction between water, inorganic particles, organic particles and pores that can exhibit heterogeneous behaviour. Therefore, flocculation, with autonomous and interactive physical, chemical and biological complex reactions, has significant implications for sediment and sediment-associated contaminants transport (Droppo, 2001). Accordingly, increase in the knowledge related to the fine sediment transportation related processes is essential to the overall understanding of sediment transport and its consequences, and therefore, to attempts to mitigate the negative effects derived therefrom.

Catchment sediment yield (t km^{-2} yr^{-1}) is "the integrated result of all erosion and sediment transporting processes operating in a catchment" (Vanmaercke et al., 2011b). However, the extrapolation of soil erosion rates to catchment sediment yields does not accurately represent the complexity and spatial variations between upstream erosive processes and sediment mobilization through hillslopes and channels (de Vente et al., 2007; Vanmaercke et al., 2011a; Walling, 1983). The difficulties of linking on-site erosion processes with sediment loss through catchment outlet was defined by Walling (1983) as the sediment delivery problem. Only a fraction of the upstream reaches the outlet in sediment form.

The sediment delivery ratio*,* is the proportion of sediment that reaches the catchment outlet (sediment yield) in relation to the quantified erosion within the catchment (gross erosion; t km^2 yr^{-1}) (de Vente et al., 2007; Maner, 1958; Walling, 1983) as:

$$Sediment\ delivery\ ratio = \frac{Sediment\ yield}{Gross\ erosion}$$

Nevertheless, the nature of the catchment, i.e. sediment source location, topographical features, channel condition and drainage patterns, vegetation type status, land use and soil texture influence the sediment delivery ratio. The influence differs for every individual catchment and results in temporal discontinuity and spatial heterogeneity in sediment transfer, generating discrepancies between the amount of soil eroded and exported at the catchment outlet. This in turn hinders the conception of a simple relationship between sediment yield and gross erosion, because the spatial and temporal lumping and its black box nature prevent the generation of generally applicable predictive rules (Walling, 1983). Technical advances (e.g. sediment tracing, connectivity indexes) in the study of geomorphological processes made possible a better understanding of source-sink relationships, the role of catchment configuration and the influence of natural (e.g. vegetation, soil texture) and human (e.g. agricultural terraces, land uses) features. For instance, sediment tracing techniques (e.g. sediment fingerprinting) and soil redistribution investigations using environmental radionuclides (e.g. ^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be) used in combination with traditional hydro-sedimentary monitoring can be used to establish integrated sediment budgets (Collins et al., 2001; Navas et al., 2014; Porto et al., 2011; Walling, 1999; Walling and Collins, 2008).

Integrated sediment budgets are represented in the form of flow diagrams that represent the eroded input/output of sediment from different defined catchment compartments to try to elucidate where the sediment is coming from, its transport pathways and transported and stored storage zones (e.g. Dietrich and Dunne, 1978; Estrany et al., 2012; Slaymaker, 2003). River systems act as “jerky conveyor belts” (Ferguson, 1981), where sediment transfer from sources to sinks is irregular over time and space. Sediment may be lost and stored in a sink or new eroded sediment may be added to the transfer process. The sediment budget construction requires a deep understanding of the four main parts of the conveyor belt: (1) sediment delivered from sources, (2) entrainment at critical shear stress, (3) transport downstream, and (4) deposition in temporary or permanent sinks (Fryirs, 2013). However, although integrated sediment budgets have been proved useful to understand erosion/deposition dynamics within catchments, they also have some limitations, as the temporal and spatial lumping of sediments (Fryirs, 2013; Walling, 1983). In a nutshell, the temporal lumping problem refers to the time resolution of the sediment balance. If the sediment budget is constructed using a long time period dataset, results integrating sediment delivery processes occurred during different events of different magnitude. It is thus not possible to elucidate if the activation of sediment transfer within a catchment is associated with low frequency and high magnitude rainfall events that cause huge erosive processes (e.g. rill gully erosion) or by lower magnitude rain events with a lesser energy processes but continuous, i.e. sheet erosion. Conversely, spatial lumping refers to the fact that total erosion and storage processes are expressed using a single number (i.e. the sediment delivery ratio). Hence, sediment budgets do not account for spatial heterogeneity in a catchment's physical configuration, which results in significantly different sediment delivery responses within a catchment (i.e. inputs, outputs and storage). Therefore, each sediment source has a unique delivery potential, and its position relative to the channel and catchment division determines the probability that the sediment contained by a specific source will be delivered to the river system.

In this context, different sediment sources within a catchment can be activated depending on predominant hydro-sedimentary drivers (Misset et al., 2019). For example, channel actuates as the main sediment source in a catchment when slope-to-channel connectivity is not effective. The reactivation of fine sediment deposited in riverbeds, sediment bars or channel banks is governed by flow rate, shear stress, or stream power (Park and Hunt, 2017). Conversely, sediment transfer can also control erosion processes on catchment hillslopes. The precipitation intensity, runoff, or mass movements drive its effectiveness. However, vegetation changes, mass movements, sediment supply exhaustion or human disturbances (e.g. check dams, agricultural terraces) can drastically alter sediment origin (Belmont et al., 2011; Grabowski and Gurnell, 2016; Vanmaercke et al., 2017).

Identification of dominant sediment sources, quantification of their relative contributions to suspended sediment loads, as well as the determination of the resistance thresholds on the driving forces that activate them, can be an essential part of assessing the factors controlling suspended sediment transport as a surrogate of erosion problems in river catchments.

1.2. Sediment delivery in Mediterranean catchments

In Europe, crop yield reduction caused by soil erosion is estimated as non-significant in global terms (Bakker et al., 2007). However, results presented considerable spatial variability between northern and southern Europe, the Mediterranean Region being considered as the most vulnerable area (Bakker et al., 2007). The Mediterranean Region has unique characteristics worldwide, strongly marked by complex relationships between natural and human, biotic and abiotic variables (Wainwright, 2009). An irregular distribution of rainfall and the hot and dry character of summers, in combination with lithological, and physiographic characteristics promote divergent responses in erosion rates and sediment yields over time and space (García-Ruiz et al., 2013; Kosmas et al., 1997; Peña-Angulo et al., 2019). In this context, a compilation of sediment yield data from 1,794 different locations throughout Europe showed that ca. 85% of the data from the Mediterranean region exceeded 40 t km^{-2} yr^{-1}, and more than 50% exceeded 200 t km^{-2} yr^{-1}, these being the highest rates in Europe together with those of mountainous areas (Vanmaercke et al., 2011b).

The Mediterranean region has been singled out as a hotspot of Global Change dynamics, especially referring to the climate and land-use change (Gates and Ließ, 2001; Paeth et al., 2017). Climate change and land-use change are stated to be the major potential drivers of erosion processes through a more intense hydrological cycle (Borrelli et al., 2020; Luetzenburg et al., 2020; O’Neal et al., 2005). Climate projections for southern Europe predict decreases in rainfall amounts, in combination with rising temperatures and high intensity rainfall episodes (Giorgi and Lionello, 2008; Stojković et al., 2014). It is estimated that rainfall erosivity is more closely related to rainfall intensity than to rainfall volume (Borrelli et al., 2020). A change in rainfall amounts, combined with dryer conditions and an increase of the intermittency and magnitude of rainfall episodes and floods associated can eventually disturb ecosystem equilibrium. As a result, geomorphological cycles could be altered, probably increasing erosive and land degradation processes (Favis-Mortlock and Guerra, 1999; Olesen and Bindi, 2002). As a result of depth economic changes, land cover changes in Mediterranean Europe are marked principally by a dichotomous increase of urban and forested areas (Catalán et al., 2008; Gates and Ließ, 2001; Pons and Rullan, 2014; Tomaz et al., 2013). Urbanization results in a reduction of soil permeability, often resulting in increased runoff ratios, higher discharge peaks and lower lag times (Sala and Inbar, 1992). Conversely, the increase in forest coverage could lead to opposite effects. Forests mainly appeared in abandoned and marginal croplands and agricultural areas, which are more prone to high erosion rates (Poesen and Hooke, 1997; Serrat and Ludwig, 2004). The growth of forest cover generates major soil protection, increasing the infiltration capacity, decreasing surface runoff generation and erosive processes (Hooke, 2006). However, the combination of forest mass growth, rising temperatures, high intensity storms and a lack of forest management can intensify wildfire risk, one of the major causes of erosion and soil degradation in Mediterranean environments (Shakesby, 2011). Denser vegetation cover and fuel accumulation can produce flammable connectivity patches in large areas along the landscapes (Moreira et al., 2011), with higher occurrence risk of Large Forest Fires (i.e. >500 ha) in fire-prone environments. The Mediterranean basin is a fire-prone environment (Pausas et al., 2008), as evidenced by the strong forest fire regime during the mid and late Holocene (Carrión et al., 2003). The climate, characterized by a hot dry summer season, is the main control factor of the pyrogeography of the Mediterranean landscapes. The native vegetation was adapted to a particular fire regime through mechanisms of regrowth and germination (Pausas and Verdú, 2005). Furthermore, some characteristics (e.g. volatile compounds, branch and leaves accumulation) of many Mediterranean pyrophyte species (e.g. *Pinus halepensis*) promote the fast spreading of wildfires to ensure their community permanence against non-adapted species (Pausas and Verdú, 2005).

The complete or partial removal of the vegetation and litter cover cause a reduction in the interception, infiltration, evapotranspiration and sediment trapping. This combined with the alteration of some physicochemical soil properties such as water repellence, structure stability, texture and particle size distribution (Certini, 2005) can disrupt channel-slope connectivity, overland flow generation and sediment yields. Many studies have also documented the increase of overland flow generation in post-fire environments (Cosandey et al., 2005; Ferreira et al., 2005; Scott and Van Wyk, 1990; Stoof et al., 2015). This is caused mainly by the reduction in vegetation cover and the increasing of soil hydrophobicity which drastically reduces the response time during rainfall-runoff events, especially during the first post-fire year (Candela et al., 2005). This scenario, together with a lower aggregate stability, increases the sediment yield in hillslopes, as well as the sediment delivery to, and sediment fluxes within river channels, which can result in irreversible soil degradation.

Under the current context of Global Change, it becomes important to implement catchment management plans so as to protect this vulnerable environment and prevent or diminish modification of its geomorphological cycles. To this end, it is necessary to monitor erosive processes and sediment transport, as well as to detect erosion hotspots.

1.3. Catchment hydro-sedimentary monitoring

Continuous monitoring of water and sediment at catchment scale makes possible the quantification of sediment loads, sediment yields, stationary patterns and assessment flood event response to different driving forces thresholds or natural and human perturbations. At catchment scale, on-site erosion effects have a measurable off-site response (i.e. sediment and water yields). Therefore, reliable long- and short-term data is essential to assess on- and off-site effects of different erosion processes (Phillips, 2010; Walling, 1983).

Traditionally, manual sampling strategies were used to estimate sediment yields. However, continuous electronic monitoring to collect high-resolution and long term data replaced these traditional techniques, allowing a more accurate characterization of hydro-sedimentary dynamics (Walling, 1988; Wass and Leeks, 1999). The continuous water and sediment monitoring makes possible the analysis of hysteretic patterns in the relationship between discharge (Q) and suspended sediment concentrations (SSC). Hysteresis in geomorphic systems is defined as a loop-like non-linear behaviour where at least two values of a dependent variable are associated with a single value of an independent variable (Phillips, 2003; Zuecco et al., 2016). A non-linear behaviour between Q and SSC is normally related with runoff generation process at hillslope and catchment scales (Camporese et al., 2014; Dooge, 2005). Hysteretic patterns change when a driving variable (e.g. rainfall, soil moisture) exceed a certain threshold. As a result, abrupt changes can occur in a response variable (e.g. Q, SSC) because different hydrological processes may become dominant. Therefore "hysteresis is the dependence of a response variable not only on the current value of a driving variable but on its past history as well" (Camporese et al., 2014). Many works analyse the hysteretic relations between two variables in hydrology: discharge is related with rainfall (e.g. Andermann et al., 2012), groundwater (e.g. Fovet et al., 2015), soil moisture (e.g. Fortesa et al., 2020), solute concentrations (e.g. Burt et al., 2015), water temperature (e.g. Blaen et al., 2012) and suspended sediment concentrations (e.g. Fortesa et al., 2021). Between Q and SSC, hysteretic analysis can reveal different patterns on sediment connectivity, indicating the activation of different catchment compartments and relating it to driving force thresholds. Likewise, changes in sediment sources can be performed in the relationship Q and SSC, providing information about its foreseeable distance from the measurement point according to the rotation direction, the shape of the loop and its area (Williams, 1989). Hysteretic clockwise loops are associated with the activation of sediment sources that are close to the measurement point, while counter-clockwise loops are associated to sediment mobilization from remote sites within a catchment (Giménez et al., 2012; López-Tarazón and Estrany, 2017; Rovira and Batalla, 2006). Several quantitative indexes have been developed to improve hysteretic classification (e.g. Aich et al., 2014; Langlois et al., 2005; Lawler et al., 2006; Lloyd et al., 2016). These indexes provide quantitative data about hysteretic loops features, allowing the comparisons at different spatio-temporal scales, detection in pattern changes, as well as correlation with other hydro-meteorological variables such as rainfall, discharge or suspended sediment concentration. However, the evaluation of hysteretic behaviour between Q and SSC has been scarcely integrated in catchment management strategies, partially due to the differential relationship between sediment and runoff depending on the scale of study (de Boer and Campbell, 1989) and the difficult interpretation of complex hysteretic loop patterns (Sherriff et al., 2016).

1.4. Sediment source fingerprinting

To reduce the negative on- and off-site effects derived from erosion and sediment transport processes and apply correct management practices, it is necessary to detect erosion hotspots within a catchment. The EU Water Framework Directive (European Community, 2000) developed an implicit assumption about the relevance of sediment monitoring to assess the role of sediments in the ecological status of water bodies (Collins and Anthony, 2008; Perks et al., 2017). Therefore, information on the nature and relative contribution of sediment sources in river systems is emerging as a key aspect when designing and implementing specific erosion control strategies.

A methodology for determining the sediment origin that has become very relevant in the last 20 years is the sediment source fingerprinting approach also known simply as sediment fingerprinting. It has been applied at different temporal scales: from the flood event (Gaspar et al., 2019; Martínez-Carreras et al., 2010b) up to determining the origin of historically deposited sediments (e.g. over the last ca. 100 years; Pulley et al., 2018). The first references using tracers to quantify and model sediment origin in catchments date back to the 1970s (Klages and Hsieh, 1975; Wall and Wilding, 1976; Walling et al., 1979). However, the incorporation of new statistic methodologies, the implementation of un-mixing models to quantify the sediment apportion and the use of new tracers has increased its use, which is reflected in the increasing number of publications every year (Collins et al., 2020; Davis and Fox, 2009; Walling, 2013).

The technique relies on the comparison of different soil properties (i.e. tracers) between samples collected in potential erosion areas and targeted suspended sediment samples collected within the fluvial network (Figure 1.1; Haddadchi et al., 2013; Walling, 2013). In an idealized conceptual framework, (1) soil particles –

comprising potential source areas within a catchment– are detached and transported during rainfall events, (2) the eroded particles are mixed during subsequent transportation to and through the fluvial system, (3) the resultant mixture is transported by rivers in form of sediment load, (4) the soil properties used as sediment tracers reflect the spatio-temporal variations of eroding source sediment contributions and (5) source and sediment tracers can be compared in order to quantitatively estimate the apportionment of sediment provenance. Thus, a required consideration when it comes to correctly apply sediment fingerprinting is that soil and sediment properties used as tracers should be representative of the main erosion sources, must be measurable and remain stable or vary in a predictable way over time and space (Motha et al., 2002).

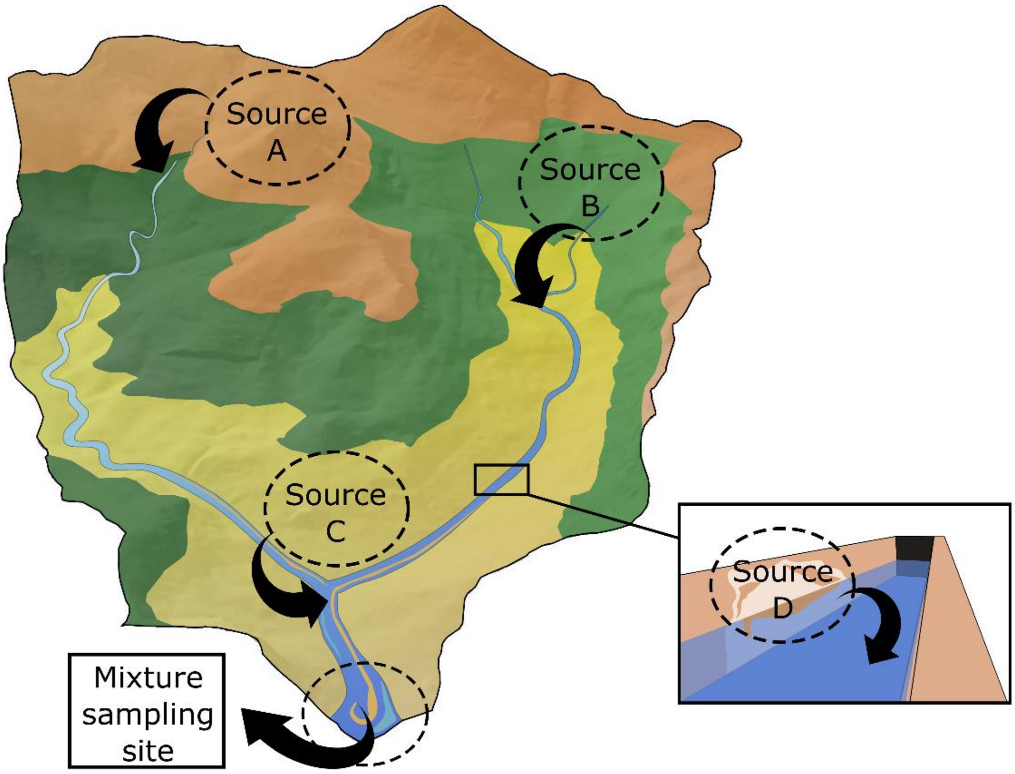


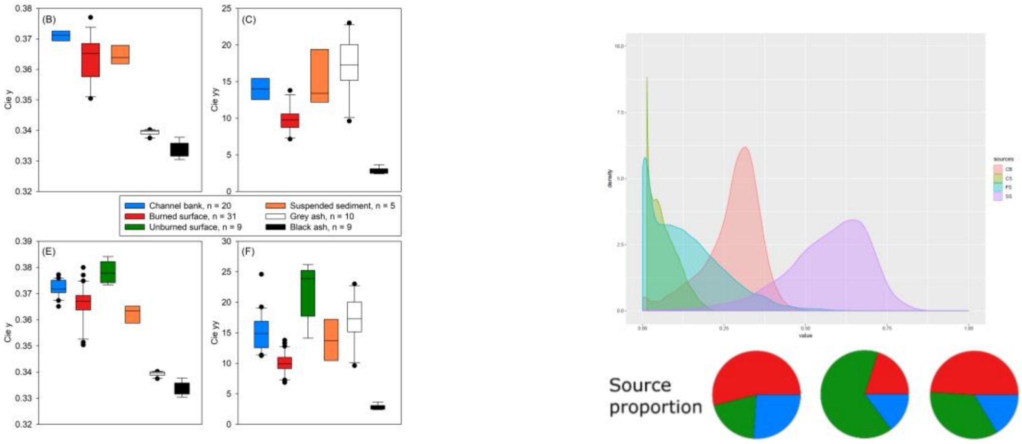
Figure 1.1. A simplified conceptual model of the sediment source fingerprinting technique based in 3 different surface sources (A, B and C; e.g. land uses, lithology) and one subsurface source (D; channel banks).

Tracers can be categorized in three different groups: (1) geochemical (e.g. inorganic elements, radionuclides and mineral magnetism), (2) biochemical (e.g. organic elements, stable isotopes, biomarkers, organic chemicals and DNA) and (3) physical (e.g. spectrometry, particle size characteristic) (Koiter et al., 2013). Therefore, several properties have been used as sediment tracers (Collins et al., 2017; Haddadchi et al., 2013), including colour parameters (e.g. Barthod et al., 2015; Grimshaw and Lewin, 1980; Martínez-Carreras et al., 2010a), grain size distribution (e.g. Kurashige and Fusejima, 1997; Weltje and Prins, 2007), clay mineralogy (e.g. Eberl, 2004; Gingele and De Deckker, 2005), mineral magnetic properties (e.g. Pulley and Collins, 2018; Yu and Oldfield, 1993), geochemistry (e.g. Chen et al., 2019; Collins and Walling, 2002), fallout radionuclide activities (e.g. Estrany et al., 2016; Evrard et al., 2020; Evrard et al., 2016; Wallbrink and Murray, 1993), cosmogenic radionuclides (e.g. Perg et al., 2003), stable isotopes (e.g. Fox and Papanicolaou, 2008), biomarkers (e.g. Reiffarth et al., 2016), pollen (e.g. Brown, 1985); and enzymatic activity (Nosrati et al., 2011).

In general, there are four key stages in the application of the sediment fingerprinting approach (Figure 1.2). The first step, sampling*,* encompasses the source classification and the source and design of the sediment sampling strategy (Figure 1.2A). There is no general protocol for soil and sediment sampling. However, a prior knowledge is required of the catchment through field observation, sediment connectivity indexes, interviews with local people or aerial photographs analysis to define erosion problem areas (Koiter et al., 2013; Krause et al., 2003; Paolo et al., 2004; Upadhayay et al., 2020). Source classification is normally done according to spatial provenance (e.g. tributary sub catchments or geological units) or source typology (e.g. surface vs. subsurface sources, land uses) depending on the objectives (Collins et al., 2017). Usually, sediment source sampling is carried out following a stratified strategy in a single field campaign. Here, it is important to collect representative samples of the source groups in areas showing evidence of slope-to-channel connectivity or erosion scars for channel bank sampling. On the other hand, the target sediment sampling for contemporary studies is usually based on suspended sediment (e.g. Navratil et al., 2012) or bed sediment (e.g. Collins and Walling, 2007) sampling. Many methods have been used to collect suspended sediment samples at the catchment outlets. Submersible pumps, auto samplers, portable continuous-flow centrifuge or manual sampling using bottles were the most common for sediment sampling during flood events (Collins et al., 2017; Davis and Fox, 2009). However, in numerous studies (Ankers et al., 2003; Estrany et al., 2016; Evans et al., 2006; Koiter et al., 2013; Laceby et al., 2015; Martínez-Carreras et al., 2010c) the authors opted to collect time-integrated sediment samples using time-integrated sediment traps (Phillips et al., 2000). For bed sediment sampling, re-suspension techniques were the most common (Duerdoth et al., 2015; Estrany et al., 2011; Lambert and Walling, 1988). Conversely, for long-term scales or historical sediment sampling, authors usually extract sediment cores from depositional zones, floodplains, reservoirs, wetlands or lake deposits (Foster et al., 2006; Li et al., 2020; Miller et al., 2005; Navas et al., 2011; Pulley et al., 2015).

(A) Sampling

(B) Sample treatment and tracer measurements



(C) Tracers accuracy analysis

(D) Sediment source ascription

Figure 1.2. General four key steps in the application of sediment source fingerprinting

The second step is the pre-processing of sediment and source samples and tracer measurements (Figure 1.2B). Samples might be treated to eventually solve some issues that influence a tracer’s conservativeness (e.g. sieving at different fractions). In this stage, researches largely addressed the influence of particle size and organic matter on tracer values (e.g. Crockford and Olley, 1998; Hill et al., 1998; Koiter et al., 2018; Laceby et al., 2017). Several ways to address particle size and organic matter differences between sources and sediment are: sieving to a specific size (e.g. He and Walling, 1996), removing the organic content (e.g. Pulley and Rowntree, 2016) and using correction factors (e.g. Collins et al., 1997). However, there may be processes relevant to a tracer's concentration stability that are independent of particle size or organic content, which need to be further investigated. Measurement techniques depend on the budget available and the tracer set selection. Although the technique has evolved with advances in computing technology and the inclusion of new tracers, some of the most used methods are the following: gamma spectrometry to measure fallout radionuclides activity (e.g. Owens and Walling., 1996), microwave digestion coupled with inductively coupled plasma mass spectrometry (ICP-MS) analysis for geochemical elements (e.g. Estrany et al., 2011), magnetic susceptibility sensors for mineral magnetics (e.g. Ramon et al., 2020) and diffuse reflectance spectrometers for colour measurements (e.g. Martínez-Carreras et al., 2010c).

At the third stage, tracer accuracy analysis (Figure 1.2C), is mainly focused on tracer conservative behaviour and its ability to discriminate between source categories. To deal with these two issues, the statistical procedure used most often was described by Collins and Walling (2002). This two-step process involves the performing of a Kruskal-Wallis *H* test to assess if the selected tracers discriminate between the different source categories, and a Discriminant Function Analysis (DFA) so as to select the optimum tracer set. However, other static procedures have been used such as Principal Component Analysis (PCA; e.g. Walling, 2005), the Mann–Whitney *U* test (e.g. Carter et al., 2003), Wilcoxon rank-sum test (e.g. Juracek and Ziegler, 2009), the Tukey test (e.g. Motha et al., 2003), *t* test (e.g. Hancock and Revill, 2013), conservativeness index and a ranking based on consensus (Lizaga et al., 2020a) or tracer-particle size relationships and source mixing polygons (Smith et al., 2018). More recently, the use of artificial mixtures with known source proportions has been introduced in fingerprint researches (e.g. Brosinsky et al., 2014) to investigate the linear additivity behaviour of tracers by comparing measured and predicted values using a mass balance approach.

Finally, step 4 is the sediment source ascription (Figure 1.2D). Quantitative estimations of the relative contributions of each source to sediment samples are assessed mathematically by using unmixing models (Collins et al., 1997; Walling and Woodward, 1995; Yu and Oldfield, 1993, 1989). Recently, Lizaga et al. (2020b) developed the FingerPro mixing model as R package, incorporating statistical and graphical tools for the selection of unmixing dataset to optimize results. Frequentist linear mixing models are normally based on the solving of a system of linear equations based on chemical mass conservation. Models are normally constrained as source type contributions sum to unity. Solutions are usually obtained by minimizing the errors associated with the system of equations, so that the differences between estimated and measured tracer values in the target samples are minimised. Additionally, other types of statistical models have expanded greatly. Bayesian mixing models (e.g. Abban et al., 2016; Blake et al., 2018; Massoudieh et al., 2013; Nosrati et al., 2014; Stock et al., 2018; Stock and Semmens, 2016), which allow a better representation of the natural variability in sources and sediment data due to their flexible likelihood-based structure. To a lesser extent, End Member Mixing Models (EMMA; Mukundan et al., 2010; Rose et al., 2018) based on performing a principal component analysis (PCA) with the tracer data measured on the sediment samples collected at the outlet, have also been used.

Despite the constant evolution and improvement of the sediment source fingerprinting approach, there are still a number of challenges to be addressed to reduce the uncertainties inherent in the technique. These uncertainties are mainly associated with sampling methodologies (e.g. Manjoro et al., 2017), spatial variability of source material properties (e.g. Du and Walling, 2017), statistical models selection and optimization (e.g. Haddadchi et al., 2014; Nosrati et al., 2014; Palazón and Navas, 2017), and alteration of soil properties during conveyance or temporal deposition within the river channel (e.g. Koiter et al., 2013). These aspects warrant further research in order to assess the degree to which the assumptions of the sediment fingerprinting approach are met. Similarly, land-use managers/regulators have a relatively poor understanding of fingerprinting techniques, and therefore do not necessarily understand the benefits of incorporating such methods into their management framework (Miller et al., 2015). In this respect, the development of economic and rapid methodologies might be essential for its application as a catchment management tool.

1.5. Sediment fingerprinting and hydro-sedimentary monitoring as tools for catchment management

Catchment/watershed management involves all human actions aimed at the manipulation of resources so as to provide goods and services without adversely affecting the ecosystem status (FAO, 1998). Catchment management has evolved into integrated catchment management, which assimilates social, technical and institutional dimensions. It can be defined as a “process that recognises the catchment as the appropriate organising unit for understanding and managing ecosystem processes in a context that includes social, economic and political considerations, and guides communities towards an agreed vision of sustainable natural resource management in their catchment” (Fenemor et al., 2011).

Earth science research is essential for catchment management. The best management practices require an understanding of how the different elements of the landscape (e.g. soil, water, land uses, human structures) interact, and recognition of the linkages between upstream and downstream processes. Source control practices are essential in catchment management. The empirically-based Universal Soil Loss Equation (USLE) and other related models (i.e. Revised Universal Soil Loss Equation [RUSLE], Modified Universal Soil Loss Equation [MUSLE], Chemicals, Runoff, and Erosion from Agricultural Management Systems [CREAMS], Groundwater Loading Effects of Agricultural Management Systems [GLEAMS] and the Water Erosion Prediction Project [WEPP]) are often used to predict erosion (Drake and Hogan, 2013). However, despite the usefulness of models, a good understanding of physical processes occurring on the soil surface and interactions between the different catchment components and sediment delivery driving forces is crucial for the development and application of the optimal erosion control measures. Therefore, it is necessary to collect reliable information about sediment mobilization through hillslopes and within the channel, and downstream sediment yields. However, a widespread adoption of standard methodologies to evaluate sediment transport dynamics and identify major sediment production areas is still missing (Du and Walling, 2017; McCarney-Castle et al., 2017; Walling and Collins, 2008). Information about spatiotemporal variation in suspended sediment sources, concentrations and loads can be assessed by a combined approach of continuous monitoring of water and sediment fluxes with sediment sources fingerprinting. In a highly variable environment such as the Mediterranean Region, collecting constant information about sediment delivery throughout the year should be considered. Furthermore, long datasets (e.g. several years) are needed in order to compute past trends and eventually account for global change. In addition, this information is essential for the development of sediment transport models that integrate information on sediment origin (Owens et al., 2005; Perks et al., 2017; Vercruysse et al., 2019).

1.6. Hypothesis and objectives

The working **hypothesis** of this thesis are:

H1: *Optimization of the sediment fingerprinting technique through research on the conservative behaviour of soil parameters and the use of low-cost and fast-to-measure tracers allowing evaluation of some of the assumptions underlying the technique, improvement of its applicability and the reduction of uncertainties.*

H2: *Hydro-sedimentary monitoring combined with sediment fingerprinting makes possible a better identification of the activation patterns of the different sediment sources within a catchment, resulting in a useful tool for catchment management.*

One general objective is proposed:

GO1: *To identify erosion and sediment transport processes (functional connectivity) in two Mediterranean catchments affected by different global change processes at different spatio-temporal scales, by improving current techniques for sediment origin determination (i.e., reducing uncertainties, time and cost) for its better implementation in catchment management plans.*

The general objective is developed within the five core chapters through the following specific objectives:

SO1: *To assess the water and suspended sediment yields and their dynamics in a Mediterranean catchment representative of terraced landscapes affected by afforestation and recurrent wildfires during the first three post-fire hydrological years (2013–2016.*

SO2: *To use* ^{137}Cs *and* $^{210}Pb_{ex}$ *radioisotopes as tracers to recognise the effect of fire on sediment sources during the first post-fire flush in a Mediterranean temporary stream three months after a severe wildfire.*

SO3: *To evaluate sediment colour parameters for predicting relative contributions of burned and unburned sources in fire-affected catchments.*

SO4: *To analyse and link sediment sources contributions with the hydro-sedimentary response of a catchment, thus determining the main factors regulating sediment source contributions and evaluating the potential of hydro-sedimentary monitoring combined with sediment fingerprinting as a catchment management tool.*

SO5: *To investigate eventual changes occurring within the most common soil properties used as tracers in sediment fingerprinting studies due to submersion and in-channel storage.*

1.7. Thesis structure

The thesis *corpus* is composed of a paper compendium divided into 9 chapters. Chapters 4, 5, and 6 correspond to research articles published in scientific journals, Chapter 7 to a manuscript under review for publication, and Chapter 8 to a manuscript in preparation for submission (Table 1.1, Figure 1.3.).

Table 1.1. Title, keywords, journal and status of the research articles of the thesis.

Chapter	Title	Keywords	Journal	Status
Chapter 4	Post-fire hydrological response and suspended sediment transport of a terraced Mediterranean catchment	Sediment delivery processes, wildfires, terraces, nested catchments, Mediterranean fluvial systems	Earth Surface Processes and Landforms	Published
Chapter 5	Source ascription in bed sediments of a Mediterranean temporary stream after the first post-fire flush	First flush sediment sources, wildfire disturbances, fingerprinting technique, fallout radionuclides, Mediterranean fluvial systems	Journal of Soils and Sediments	Published
Chapter 6	Analysis of post-fire suspended sediment sources by using colour parameters	Sediment fingerprinting, colour, fallout radionuclides, wildfire, ash, suspended sediment sources	Geoderma	Published
Chapter 7	Combining sediment fingerprinting and hydro-sedimentary monitoring to assess suspended sediment provenance in a mid-mountainous Mediterranean catchment	Sediment fingerprinting, End Member Mixing Analysis, hysteresis, hydro-sedimentary dynamics, sediment sources, Mediterranean catchments	Journal of Environmental Management	Under review
Chapter 8	Preliminary results: In-channel alterations of soil properties used as tracers in sediment fingerprinting studies	-	-	In preparation

Chapter 1 is a general introduction that briefly reviews the state of the art of erosion processes, the role of hydro-sedimentary monitoring and sediment fingerprinting to mitigate negative impacts promoted by the soil loss and sediment transport and their potential as management tools.

Chapter 2 is an overall description of the thesis study areas.

Chapter 3 details the methods mainly used to achieve the established scientific objectives.

Chapter 4 is related to the specific objective 1. Runoff and suspended sediment transport dynamics and its post-fire evolution are analysed in a fire affected Mediterranean catchment.

Chapter 5 is related to the specific objective 2. In this chapter ^{137}Cs and ^{210}Pb$_{ex}$ were used to quantify the relative contribution of different sediment sources to the fine bed sediment temporarily stored on the riverbed surface during the first post-fire flush.

Chapter 6 is related to the specific objective 3. Here, sediment colour and fallout radionuclides are used to distinguish between burned and unburned sources in a fire affected catchment.

Chapter 7 is related to the specific objective 4. In this chapter, two different source apportion models were evaluated. After that, changes in source contributions were linked to the hydro-sedimentary response of different magnitude flood events to determine the main factors regulating sediment contributions.

Chapter 8 is related to the specific objective 5. Here the conservative behaviour of different properties of the sediment was evaluated after a one-year experiment.

Chapter 9 contains a general discussion and conclusions.

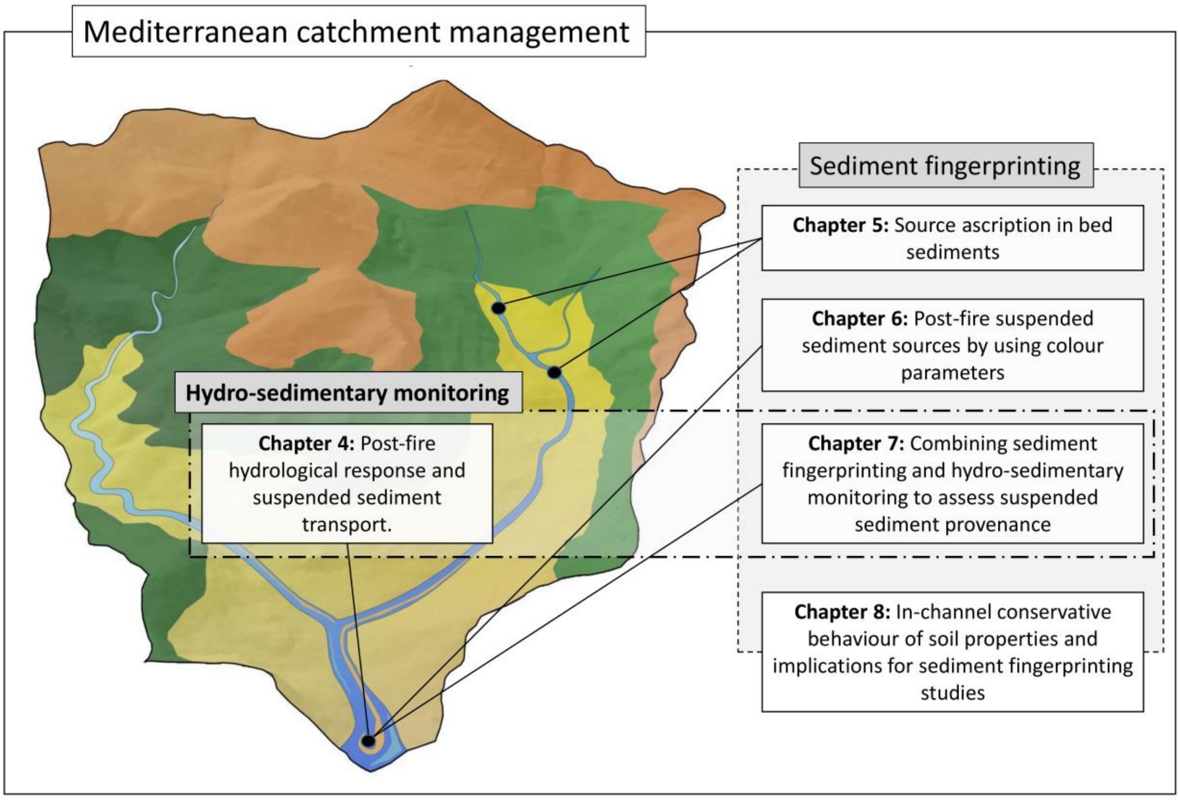


Figure 1.3. Links between chapters that compose the thesis paper compendium. The chapter titles have been shortened for clarity in the figure.

1.8. References

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2. Study areas

2.1. Overview

Located in the western Mediterranean Sea, Mallorca is the largest island of the Balearic archipelago with an area of 3620 km^2 (Figure 2.1A). The island's morphological structure was determined by its genesis, generated by the compressive stresses which occurred during Alpine orogeny in the Upper Cretaceous. Materials from the Carboniferous to the Middle Miocene were raised above sea level forming the principal mountainous reliefs, while Pliocene and Quaternary materials filled the distensile trenches of the Central Plain and the south-eastern coast (Álvaro et al., 1989). As a consequence, the island is dominated by a SE to NW horst-graben structure that compose its principal structural units: the Llevant Ranges, the Campos and Manacor basins, the Central Ranges, the Palma, Inca and Sa Pobla basins, and the Tramuntana Range (Figure 2.1B).

This thesis is focused on two small catchments < 5 km^2 (the Sa Font de la Vila and Es Fangar creek catchments) located in the Tramuntana Range. The study areas are representative of the natural and human Mediterranean mountainous landscapes, where the complex reliefs strongly determined human occupation, land uses and land management practices. In addition, the Sa Font de la Vila catchment was affected recurrently by forest fires, including the largest fire registered in the Balearic Islands.

The Tramuntana Range is the most abrupt horst of the island of Mallorca. It is located along the NW coast of the island, in a SE-NW direction (Figure 2.1B). It has a total length of ca. 90 km and a maximum width of ca. 20 km, occupying approximately an area of 1,000 km^2. Its highest point is the Puig Major peak with an altitude of 1,445 m.a.s.l. The Tramuntana Range structure is mainly composed of Mesozoic materials in the form of thrusts on Tertiary deposits. The predominant lithologies were Jurassic micrite limestone’s, limestone conglomerates and calcarenites of the Lower Miocene superimposed on clays of the Upper Triassic (Keuper; Figure 2.1C). According to the Emberger classification, the southern part of the Tramuntana Range shows a Mediterranean warm sub-humid climate, while the central and eastern parts were classified as Mediterranean humid and super-humid (Guijarro, 1986). Average annual rainfall ranges from 1,200 mm yr^{-1} to 700 mm yr^{-1} (Figure 2.1D) with a maximum rainfall in 24 hours for a return period of 25 years from 110mm in the western part to 250 mm in central areas.

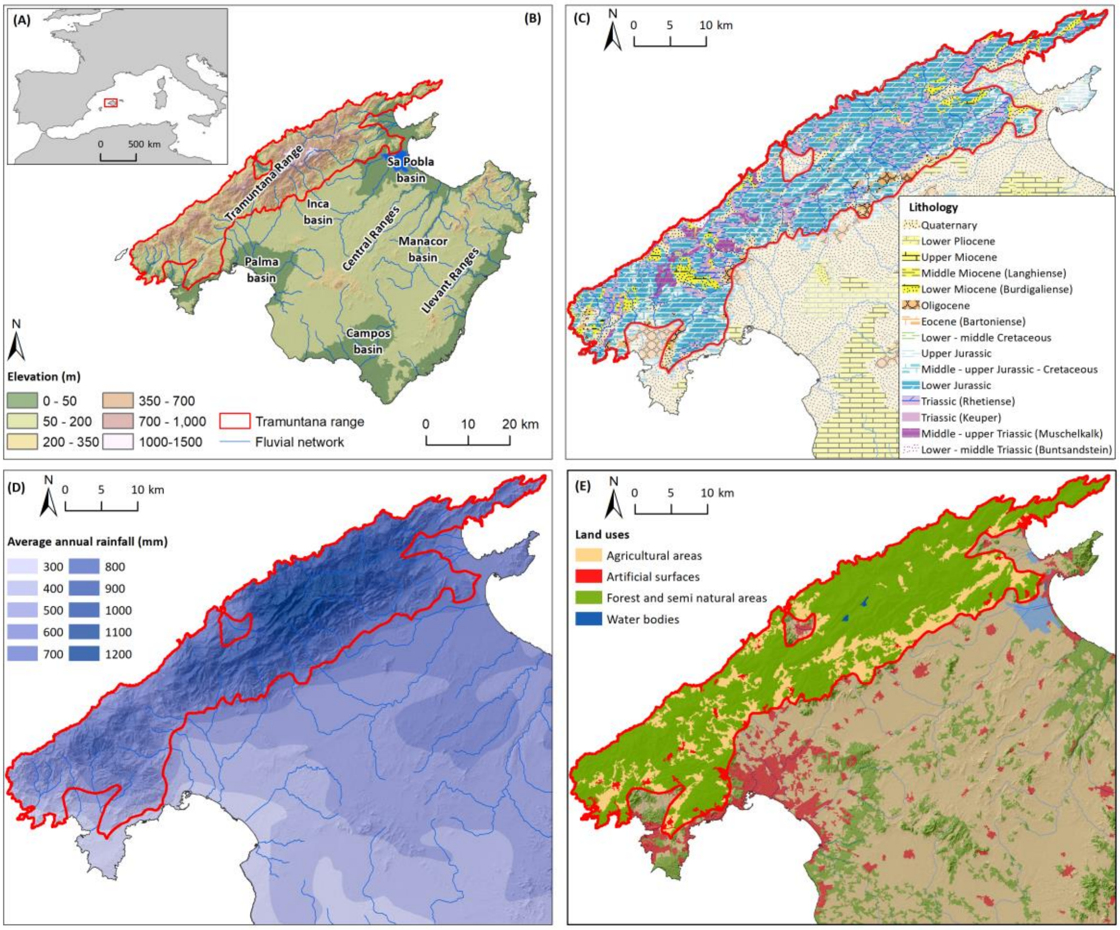

Figure 2.1. (A) Location of Mallorca Island in the Western Mediterranean Sea basin. (B) Physical characteristics of Mallorca Island and Tramuntana Range location (C) Tramnuntana Range lithology (D) rainfall distribution and (E) land use distribution.

The Tramuntana Range was occupied and exploited by humans during centuries. The landscape has been modified for agricultural use through the construction of dry stone agricultural terraces, check dam terraces and other human structures. However, the economic changes which occurred during the second half of the 20th century caused the depopulation of rural areas and a gradual abandonment of an agriculture-based economy. As a consequence, afforestation processes in former croplands caused an increase of the forest area estimated in Mallorca at 79% between 1971 and 2010 (MAGRAMA, 2012). Land use distribution according to Corine Land Cover 2018

(European Evironment Agency-EEA, 2018) were 72% forest and semi natural areas, 23.4% agricultural areas, 3.4% artificial surface and 0.6% water bodies (Figure 2.1E).

2.2. Sa Font de la Vila River catchment

The Sa Font de la Vila River is a Mediterranean catchment of 4.8 km^2 located in the Andratx municipality (western Mallorca, Spain; Figure 2.2A and 2.2B), which is affected by extensive afforestation of former agricultural land and recurrent wildfires. Catchment lithology in bottom valleys consists mainly of Upper Triassic (Keuper) clays and loams on gentle slopes (ca. < 10 degrees). Rhaetian dolomite and Lias limestone predominate in the upper parts with steeper slopes > 30% (Figure 2.2C). Soils are classified as *BK45-2bc*, corresponding to *Calcic Cambisols* (Jahn et al., 2006). The fluvial network consists of two main streams: (a) Sa Coma Freda (east, 2.3 km^2), which has a significant groundwater influence with several karstic springs; and (b) Can Cabrit (west, 2.08 km^2), which is not affected by this groundwater influence due to the reduced presence of impervious materials. In addition, a check-dam was built at Can Cabrit in 2007 (5 m high and 16 m long; Figure 2.2D).

The climate is classified as Mediterranean temperate sub-humid at headwaters and warm sub-humid at the outlet (Emberger climatic classification; Guijarro, 1986). The average temperature is 16.5°C. The mean annual rainfall is 518 mm yr^{-1}, with an inter-annual coefficient of variation of 29%. High-intensity rainstorms with a recurrence period of 10 years may reach 85 mm in 24 hours (1974-2010; data from the B118 S'Alqueria meteorological station of the Spanish State Meteorological Agency (AEMET); Figure 2.2B).

In recent decades, the Sa Font de la Vila catchment has been affected by major wildfires in 1994 and 2013 (Figure 2.2E). Before the 2013 wildfire, the catchment was mainly covered by natural vegetation (84%; Figure 2.2C): 51% forest and 33% scrubland. The rest of the catchment was covered by rain-fed tree crops (12%), rain-fed herbaceous crops (1%) and urban uses (3%). Traditional soil and water conservation structures (i.e., hillslope and valley-bottom terraces) cover 37% of the total surface area (Figure 2.2D).

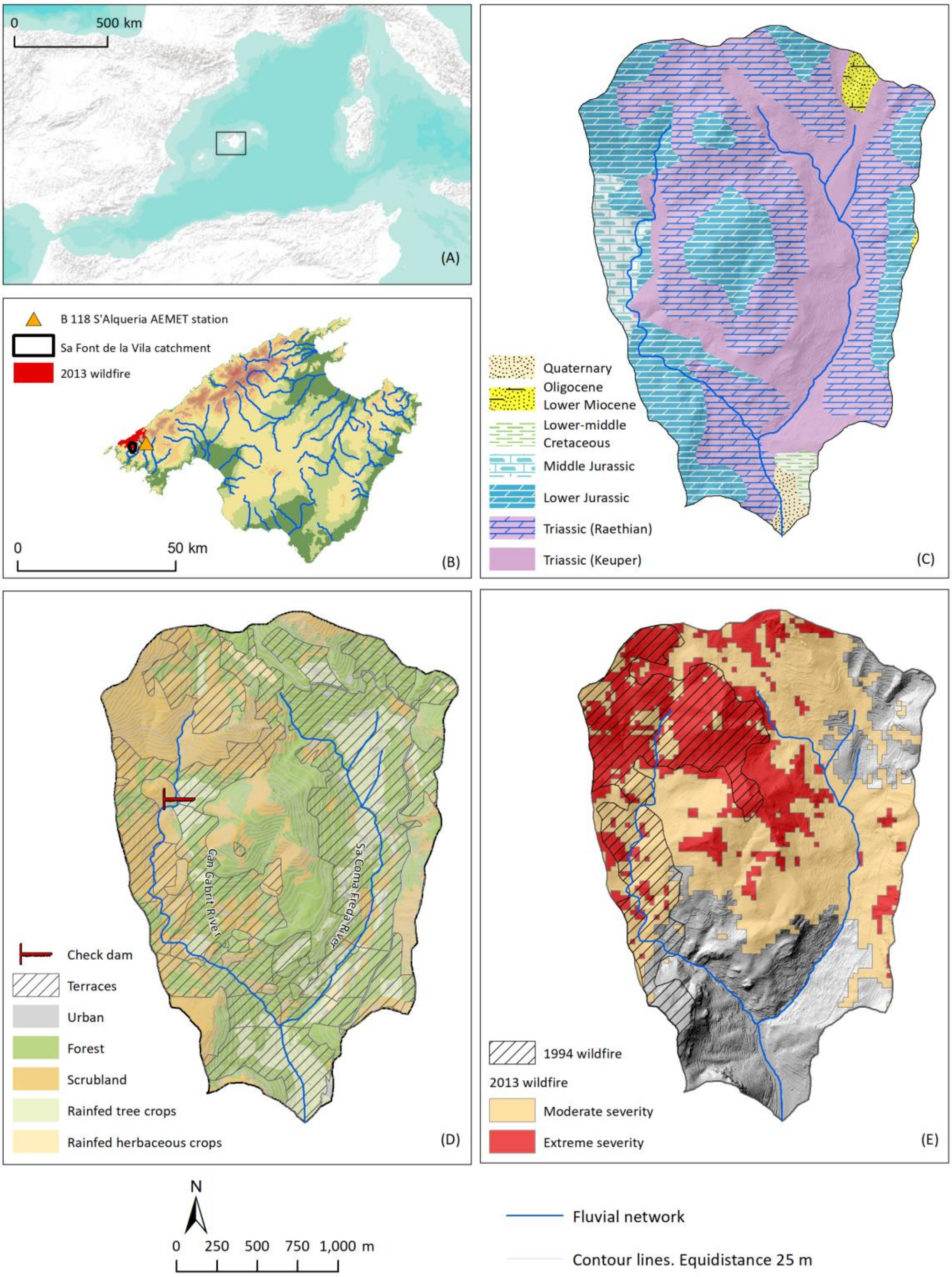

Figure 2.2. Location of the Mallorca Island within the Mediterranean Sea (A); location of the Sa Font de la Vila catchment, the area affected by the July 2013 wildfire, the B'12 S'Alqueria meteorological station and the village of Llucmajor (B); lithology (C) land uses and soil conservation practices (D) of the Sa Font de la Vila catchment (downstream site) and Sa Murtera sub-catchment (upstream site); and 1994 and 2013 wildfire affected areas as well as severity of the 2013 wildfire and 2016 sampling area (E). Channel bank and surface sampling points indicated as blue dots and orange squares, respectively

Its abandonment and degradation, involving the collapse of dry-stone walls, increased the sensitivity of the catchment (Calsamiglia et al., 2017). Collapses were higher on those abandoned terraces affected by recurrent fires due to soil degradation (Lucas-Borja et al., 2018).

The 1994 fire affected 45% of the catchment surface, whereas the 2013 one reached 71% (more than half of it had already been burned in 1994). A severity assessment with the Normalized Burn Ratio (Escuin et al., 2008) and Landsat 8 images for the 2013 wildfire assigned high and moderate severity to 24% and 47% of the catchment, respectively (Bauzà, 2014. Figure 2.2E). In addition, after the 2013 wildfire the Balearic Islands Department of the Environment (Conselleria de Medi Ambient, Agricultura i Pesca) implemented a series of post-fire strategies to prevent soil loss and degradation, which included mulching, tree planting and the creation of log barriers with dead biomass.

2.3. Es Fangar Creek catchment

Es Fangar catchment (3.4 km^2; Figure 2.3A and B) is located at the north-east side of the Tramuntana mountain range in Mallorca (Spain). Altitudes range between 404 m.a.s.l. and 72 m.a.s.l. with an average slope of 26%. The lithology is composed of massive calcareous and dolomite materials from lower Jurassic and dolomite and marls formations from the Triassic (Rhaetian) in the upper parts, being Jurassic and Cretaceous marl-limestone’s in the valley bottoms (Figure 2.3C). As a consequence of its geologic structure, and the high water availability at field capacity of the valley soils, the catchment has had an intensive agricultural activity in the past. The stream network is natural at the upper parts, however, at the valley bottom the main channel was diverted and constricted by dry stone walls for better agricultural land exploitation. Check-dam and agricultural terraces were built to control torrential floods and avoid soil erosion (Figure 2.3C and D). In addition, subsurface tile drains were built in crop lands to promote drainage, avoiding soil saturation during the wet season. 16% of the catchment is occupied by soil and water conservation structures, which means 32.4 km of dry stone walls. Nowadays, land use occupation is forest (47%), rainfed herbaceous crops fields (36%) and scrubland (17%).

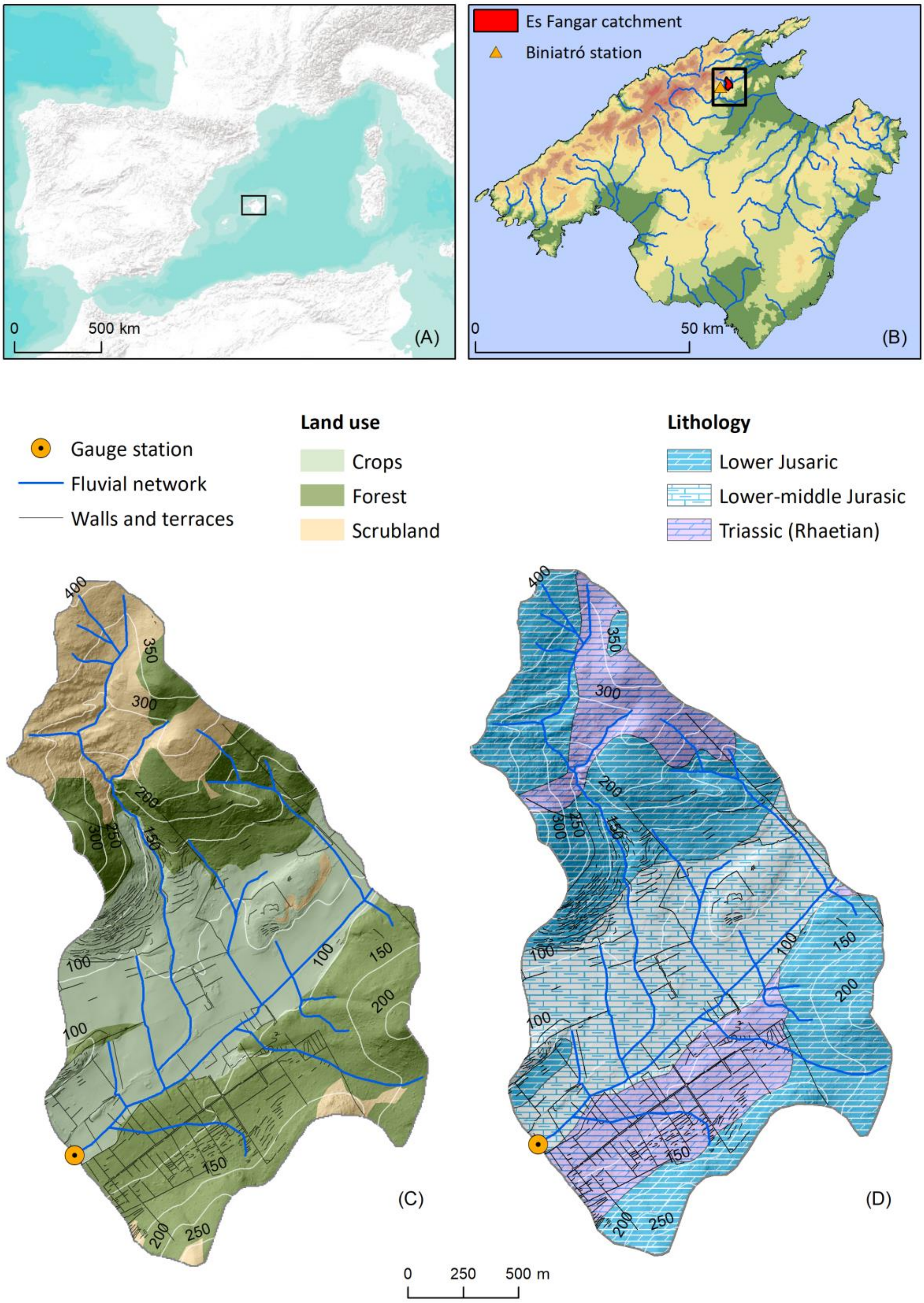

Figure 2.3. (A) Map showing the location of Mallorca in the Western Mediterranean. (B) Location of Es Fangar catchment within Mallorca island. Drainage network and terraced areas over (C) land use and (D) lithology maps.

Since the 1950s, as a consequence of an economic transition from the primary to the tertiary sector, the agricultural area was reduced by a ca. 17%. Afforestation processes have occurred in abandoned fields currently being covered by forests. According to the Emberger classification, the climate is Mediterranean temperate sub-humid (Guijarro, 1986). Mean annual rainfall is 926 mm yr^{-1} (1964-2017, Biniatró AEMET station, 1.1 km west from the study area) with a 23% coefficient of variation and the average annual temperature is 15.7 °C. Rainfall amount estimation of 180 mm in 24 hours in a recurrence period of 25 years. The hydrological regime is categorized as intermittent flashy (49% of zero days; Fortesa et al., 2020a). The annual runoff coefficient ranged from 2.9% to 14.2% (average of 10.4%) and quick flow contribution from 9.9% to 45% (average of 33%) illustrating a huge intra-annual variability of the rainfall-runoff relationship (Fortesa et al., 2020a). 80% of the sediment load is exported during autumn and winter, with an annual average sediment yield of 5.38 t km^{-2} y^{-1}.

2.4. References

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3. Methodology

3.1. Overview

In this chapter it is presented a summary of the main methods used in Chapters 4, 5, 6, 7 and 8. It is possible to divide in three different groups the principal methodologies used to reach the scientific objectives proposed (Figure 3.1). First group, continuous water and sediment monitoring (Figure 3.1A), encompass all methods related to hydro-sedimentary data acquisition and treatment, hysteretic loop analysis and detection of the main driving forces that control suspended sediment transport on the study areas (see section 3.2). Second and third groups are related with Sediment source fingerprinting. Within the second group are listed (Figure 3.1B) all steps followed to recognize main sediment sources in the study areas, including different source categories consideration, tracer's sets, tracer accuracy tests and statistical apportion models (see Chapter 3.3). Finally, the third group (Figure 3.1C) explain the methodology followed in Chapter 8 to perform the experiment on sediment properties conservative behaviour (see section 3.4).

3.2. Continuous water and sediment monitoring

Gauge stations were installed in the outlets of the two study areas (i.e. Sa Font de la Vila and Es Fangar catchments) to continuously monitor water and suspended sediment fluxes (Figure 3.2 and 3.3).

In Sa Font de la Vila catchment (Chapter 4, 5 and 6), a continuous water and sediment yield monitoring programme was implemented by instrumenting two nested gauging stations (Figure 3.2), Sa Murtera (an upstream sub-catchment; 1.1 km^2) and Sa Font de la Vila (closing 4.8 km^2). Sa Murtera gauging station is located in the northeast headwater area of the Sa Font de la Vila catchment (Figure 3.2A). The monitoring station was installed in a place where channel banks consist of dry-stone walls working as a control section for higher discharges and another smaller section built for measuring base flow.

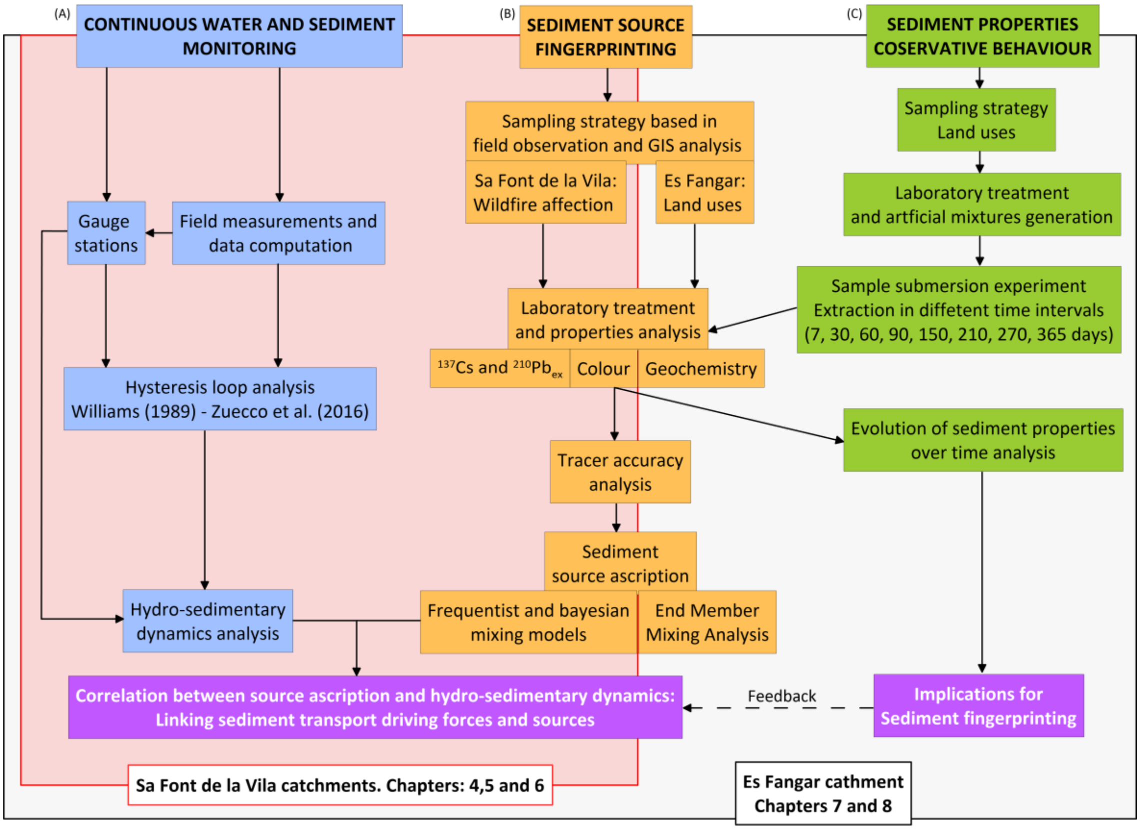

Figure 3.1. Methodological workflow encompassing main methodologies used for (A) continuous monitoring of water and sediment, (b) Sediment source Fingerprinting and (C) experiment on the conservative behaviour of sediment properties. Inside the red square are listed the methods applied in Sa Font the la Vila catchment and inside the black square the methods applied in Es Fangar catchment.

The gauging station was equipped with a Campbell Scientific CR200X data logger that stored the average values of water surface level and turbidity, based on 1-minute readings at 15-minute intervals collected by a Campbell Scientific CS451-L pressure probe and a OBS-3+ turbidimeter with a double measurement range of 0-1,000/1,000-4,000 NTU. Additionally, a rising-stage sampler modified from Schick (1967), with seven sampling bottles, was installed to provide more information on suspended sediment concentrations (SSCs). There is a 12 cm height gap between each bottle, totalling a 100 cm stage, with the first bottle located 21 cm above the riverbed. Additionally, a Casella tipping bucket rain gauge was installed.

The Sa Font de la Vila catchment gauging station was also installed in a place where channel banks consist of dry-stone walls working as a control section (Figure 3.2B). The instruments were the same as for the Sa Murtera gauging station. However, the rising-stage sampler was set up with twelve bottles, totalling a 200 cm stage.

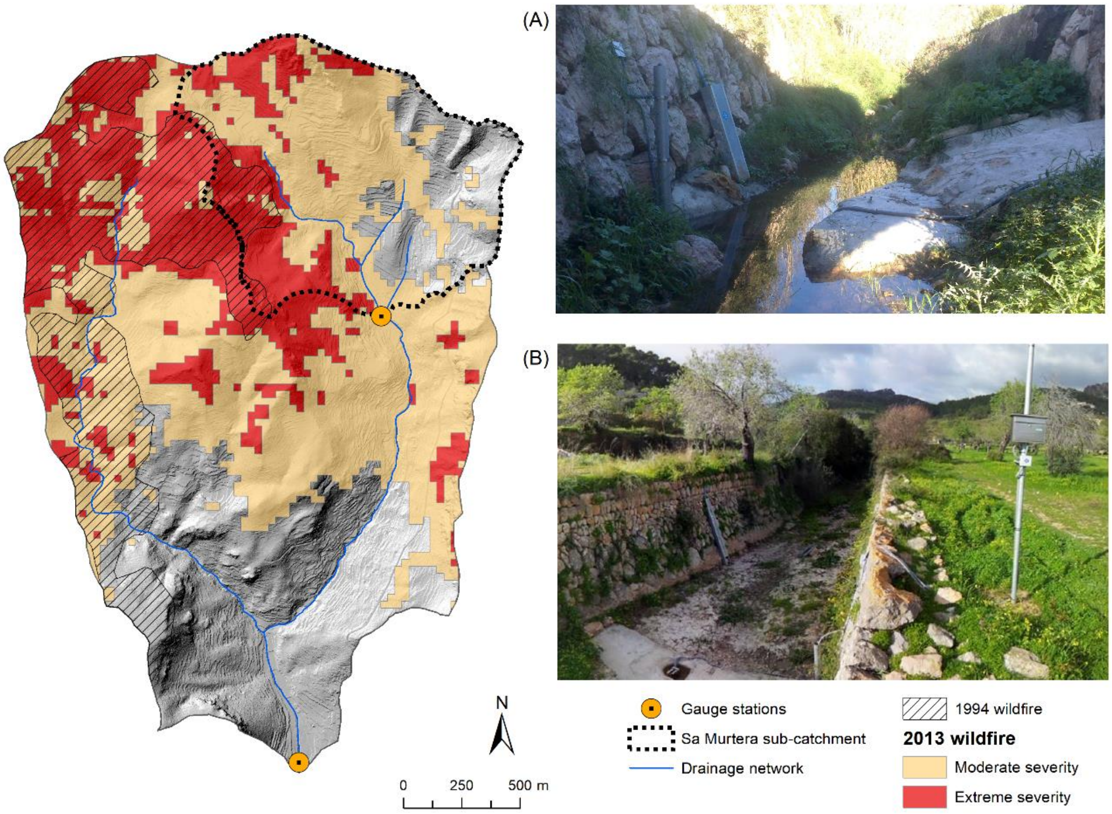

Figure 3.2. Left, Map of Sa Font the la Vila catchment with the area affected by the July 2013 wildfire, the delimitation of Sa Murtera sub-catchment and the gauge stations locations. (A) Upstream view of Sa Murtera cross section and gauge station. (B) Upstream view of Sa Font de la Vila cross section and gauge station.

In Es Fangar Creek catchment, a gauging station was installed in 2012 to continuously monitor water and suspended sediment fluxes (Figure 3.3). The station was equipped with a Campbell CS451 pressure probe and an OBS-3+ turbidimeter with a double measurement range of 0-1,000/1,000-4,000 NTU**.** Campbell CR200X logger stored 15-minutes average values of water stage and turbidity (based on 1-minute readings). In addition, in October 2014 a tipping bucket pluviometer was installed at 500 m.a.s.l. and located ca. 2.5 km away from the Es Fangar gauging station. The rain gauge was installed 1 m above the ground and connected to a HOBO Pendant G Data Logger - UA-004-64 recording rainfall at 0.2 mm resolution.

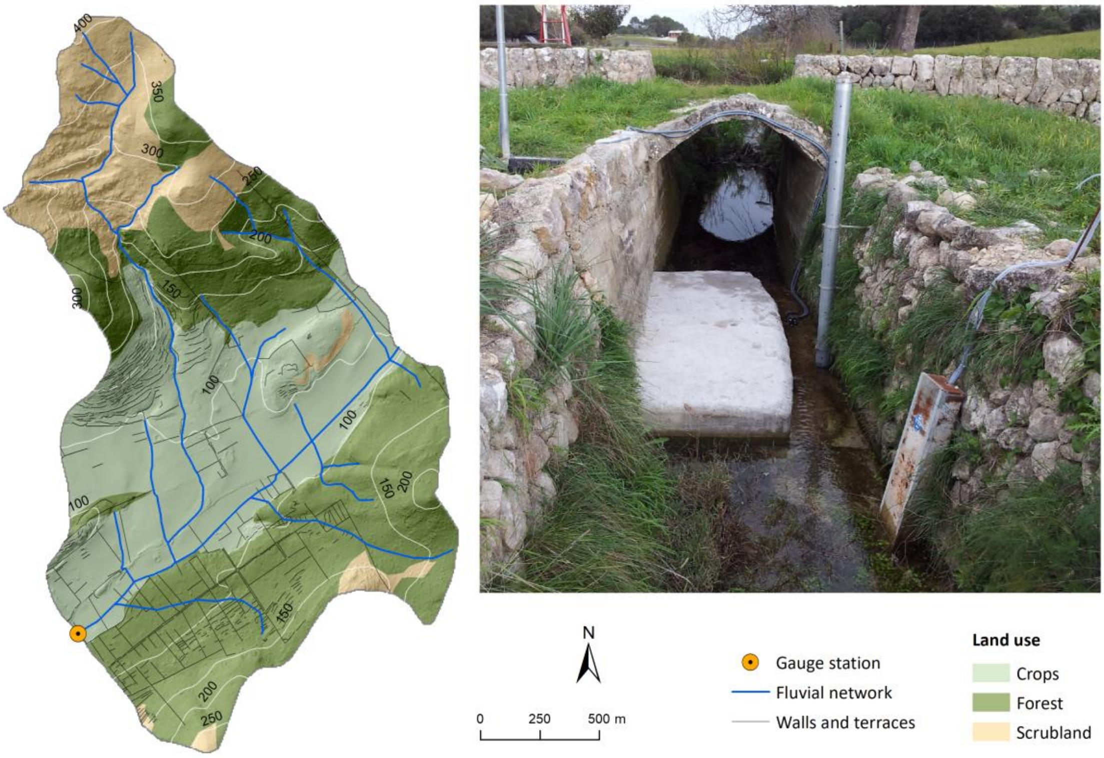

Figure 3.3. Left, Map of Es Fangar catchment with main land uses and the gauge stations location. Right, Upstream view Es Fangar cross section and gauge station.

3.2.1. Field measurements and data computation

In the three gauge stations, stream discharge was measured by an OTT MF pro inductive magnetic flow meter, with a measuring range of 0 to 6 m s^{-1} ± 2% accuracy, to develop and fit stage/discharge rating curves.

Manual depth-integrated suspended sediment samples were consistently collected (n= 55) during storm events and low flows. Samples were collected at the same sections on which turbidity probes and rising-stage samplers were installed. All the water samples were filtered by 0.45 µm cellulose esters; the filters were subsequently dried at room temperature and weighed on high-precision scales to determine the suspended sediment concentrations.

Turbidity probes were calibrated with commercial turbidity standards to check their long-term stability. The turbidity data were converted to a continuous record of SSCs by a site-specific concentration/turbidity calibration relationship. The SSCs used for calibration were measured in samples collected both manually and by rising-stage samplers. Suspended sediment loads were calculated by combining the records of SSCs provided by the turbidity sensors with the continuous records of water discharge.

The kinetic rainfall energy was calculated by the equation described by Brown and Foster (1987):

$$e = 0.29[1 - 0.72\,exp(-0.05i)]$$

where *e* is the kinetic energy of 1 mm of rainfall expressed in Mj ha^{-1} mm, and i is the rainfall intensity expressed in mm hr^{-1}. The rainfall erosivity (R) was determined by multiplying the kinetic energy of each event and the maximum intensity attained in 30 minutes (I30). The results were expressed in MJ mm ha^{-1} h^{-1}.

$$eI_{30} = R$$

Finally, the spatio-temporal relationship between discharge and sediment transport at the event scale was analysed by means of hysteresis loops applying analysis applying the classification developed by Williams in Chapter 4 (Williams, 1989) and the *h* index developed by Zuecco et al. (2016) in Chapter 7.

3.3. Sediment source fingerprinting

3.3.1. Soil and sediment sampling

A stratified sampling strategy after field observation and GIS analysis was planned in the Sa Font de la Vila and Es Fangar catchments to collect soil samples from main sediment sources.

In Sa Font de la Vila source categories were burned surface soil surface (n= 31), unburned surface soil (n= 9) and channel banks (n= 20). However, in Es Fangar catchment main land uses were considered as main sources (i.e. forest (n= 5), crop (n= 6), scrubland soils (n=5) and channel banks (n= 16)). All samples were collected in areas with visual evidence of erosion. Surface soil samples were composed by four subsamples (0-2 cm depth) collected inside a ca. 3 m radius circular area in order to include the spatial variability of the soil properties, whereas each channel bank sample was composed by three subsamples collected in a 10 m transect.

In addition, five bed sediment samples were collected along the main channel bed stem only in Sa Font de la Vila. The samples were collected a week after of high intensity storm occurred in 29th October 2013, considering the topographical characteristics of the main stem (see longitudinal profile of inlet in Figure 5.2, Chapter 5) and the wildfire's effects. During the bed sediment sampling campaign all main system channel was completely dry. Each sample consisted of two integrated subsamples, collected in the most superficial layer (ca. 5 mm) inside a heterogeneous circular area, depending on the surface of each pool.

Two parallel time integrated sediment trap samplers (Phillips et al., 2000) were installed over the channel bed in every gauge station (i.e. Sa Font de la Vila, Sa Murtera and Es Fangar) to collect time integrated suspended sediment samples, n= 4 in Sa Murtera, n= 5 in Sa Font de la Vila and n= 13 in Es Fangar.

3.3.2. Laboratory treatment and analysis

Soil and sediment samples were oven-dried at 40°C, disaggregated using a pestle and a mortar and sieved to <63 µm to minimize the differences in particle size composition between source/target samples (Walling et al., 1993). The particle size distribution and the specific surface area in source and sediment samples were determined using a Malvern Mastersizer 2000 (Chapters 5 and 6) and 3000 (Chapters 7 and 8). The Shapiro-Wilk ($p < 0.05$) normality test, Mann-Whitney *U* test and the Wilcoxon signed-rank test were applied to determine the particle size distribution similarity between each source group (soil samples) and target samples (bed and suspended sediment samples).

After the pre-treatment, each sample was closed tightly and left for more than 21 days before activity measurement, to ensure that secular equilibrium had been reached. The atmospherically-derived $^{210}Pb_{ex}$ concentration was determined by subtracting the ^{226}Ra-supported $_{210}Pb$ concentration from the total $_{210}Pb$ concentration, as $[_{210}Pb_{ex}] = [_{210}Pb] - 0.8\ [^{226}Ra]$, including a commonly used value for the reduction factor to take into account the radon emanation coefficient of soils. The ^{137}Cs, ^{226}Ra (via ^{214}Bi at 609.3 keV) and total ^{210}Pb

activity concentrations (Bq kg^{-1}) were measured by gamma spectrometry at the *Environmental Radioactivity Laboratory* of the University of the Balearic Islands using a high-purity coaxial intrinsic germanium (HPGe) detector, cooled by liquid nitrogen, shielded by 15 cm of low-background iron and equipped with high-voltage power supply, preamplifier, amplifier and multichannel analyser as an interface to a PC-type computer. The system was calibrated by a soil standard containing ^{210}Pb provided by Exeter University and a CG2-standard (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr and ^{88}Y) prepared and certified by the *Centre for Energy, Environment and Technology Research* (CIEMAT, the Spanish National reference for nuclear physics magnitudes), thus achieving a useful energy range from 25 keV to 10 MeV with a resolution of 5 keV and a detection efficiency of 0.99% for ^{137}Cs, 1.10% for $_{226}Ra$ and 4.63% for ^{210}Pb. The minimum detectable activities have been of the order of 1 Bq kg^{-1} for ^{137}Cs, 2 Bq kg^{-1} for ^{226}Ra and 12 Bq kg^{-1} for ^{210}Pb, and the uncertainties of the measurements less than 10%. As the same geometry was used for the standards and samples (less than 100 g in a vessel wide enough to assume that there are no self-absorption effects), there was no need to apply any correction factor.

Total carbon (C) and nitrogen (N) were measured by high-temperature combustion using a TruSpec CHNS, LECO. Diffuse reflectance was measured in a dark room by a spectroradiometer (ASD FieldSpect-II) at 1 µm steps over the 400-2500 µm range. The spectrometer was located in a tripod perpendicular to a flat surface, at 10 cm from the reference standard panel of known reflectivity (Spectralon). The soil and sediment samples were placed in transparent P.V.C. round petri dishes (4.7 cm diameter; Pall Corporation) and carefully smoothed with a spatula to minimize micro shadow effects due to surface roughness. The samples and the Spectralon were illuminated at an angle of 30º by a 50-w quartz halogen lamp placed at ca. 30 cm of distance. Following the International Commission on Illumination (CIE, 1931), CIE xyY colour coefficients were computed (i.e. cie x, cie y and cie yy) from the spectra reflectance measurements and the RGB colour values (i.e. red, green and blue). Then, the ColoSol software, developed by Viscarra Rossel et al. (2006), was used to estimate the Munsell HVC (i.e. Munsell H, Munsell V and Munsell C), CIE XYZ (i.e. cie X, cie Y and cie Z), CIE LAB (cie L, cie a* and cie b*), CIELUB (i.e. cie L, cie u* and cie v*), CIELHC (i.e. cie L, cie H and cie C) and decorrelated RGB (i.e. HRGB, IRGB and SRGB) colour parameters, as well as the redness index (i.e. RI) and Helmholtz chromaticity coordinates (i.e. DW nm, Pe %).

In addition, samples were placed in transparent plastic bags (7 * 5 cm) and scanned with an office scanner (Konika Minolta bizhub C554e; e.g. Krein et al., 2003; Pulley and Rowntree, 2016). The instrument was not calibrated. Red, green and blue colour parameters (i.e. RGB model) were extracted from the scanned images using *GIMP 2* open-source image-editing software. Then, the procedure using ColoSol software described in the previous paragraph was applied to convert the red, green and blue into other colour parameters.

Samples were digested according to the microwave digestion USEPA 3051A method, as follows: Pulverized soil samples (0.5 g) were transferred into polytetrafluoroethylene tubes, where 9 ml of HNO_3 and 3 ml of HCI, of high analytical purity, were added. Samples were placed in a microwave oven (Multiwave GO, Anton Paar, Austria) for 5 minutes on the temperature ramp, the necessary time to reach 175 °C; then this temperature was maintained for an additional 10 minutes. After digestion, all extracts were transferred to 100 ml flasks, filling with ultrapure water (Millipore Direct-Q System) and were filtered through 0,45 µm nylon filters (Labbox Labware, S.L). High-purity acids were used in the analyses (PamReac ApplyChem, SLU). Glassware was cleaned and decontaminated in a 10% nitric acid solution for 24 hours and then rinsed with distilled water. Calibration curves for metals determination were prepared from standard 1,000 mg l^{-1} (Sharlau, Spain). The concentrations of metals in the extracts were determined by ICP-AES (DV Optima5300, Perkin Elmer®, Inc.) equipped with a GemCone pneumatic nebulizer for viscous solutions and solutions with high content of dissolved solids (Waltham, MA, USA).

3.3.3. Particle size correction

Particle size can affect tracer concentrations on soil and sediment samples (Laceby et al., 2017). For ^{137}Cs and ^{210}Pb$_{ex}$, higher activity concentrations were observed in the fine particle size fractions (He and Walling, 1996) due to the increasing of the specific surface area (hereafter SSA, $m^2\ g^{-1}$) in these fractions (Horowitz, 1991; Rawlins et al., 2010). In Chapter 5, to minimize the possible element concentration variations generated as a result of particle-size distribution between source and target sediment samples, size fractionation to 63 µm was combined with correction procedures.

Particle-size correction factor applied in Chapter 5 was:

$$C_c = \left(\frac{S_x}{S_s}\right) C$$

where C is the measured mean property concentration in source material, C_C the property concentration corrected for particle size using SSA, S_x the SSA of suspended or deposited sediment collected at each location x, and S_S is the mean SSA of the source to be corrected. Accordingly, the particle-size correction factor was applied to those source sample groups that showed significant statistical differences with each other and with the targeted bed sediments, to avoid errors in tracer concentrations caused by the differential tracer adsorption of the finest particles (Smith and Blake, 2014).

3.3.4. Tracers accuracy

Artificial mixtures were made from 2, 3 and 4 different source samples (Chapter 6 and 7). In addition, and to investigate ash influence on the colour parameters in Chapter 6, 18 artificial samples were created mixing suspended sediment and ash (black and grey) in different proportions. The ash proportion was gradually modified from 10% to 90% to observe the influence of ash on sediment colour variation.

The individual accuracy and linear additivity behaviour of some tracers were assessed by comparing measured values and predicted values by means of a mass balance approach in artificial mixtures (i.e. tracer values in the mixture are equal to the sum of contributions from each artificially mixed sample; Chapter 6 and 7).

Kruskal-Wallis H tests were performed to assess source discrimination potential of the selected tracer’s. Discriminant Function Analysis (DFA) checked the discriminatory potential of each tracer group (taking selected tracers as independent variables) and calculated the percentage of correctly classified samples (leave-on-out cross-validation).

Range tests were used to exclude potentially non-conservative tracers in each individual suspended sediment sample. Therefore, the tracers in suspended sediment and artificial mixtures that showed values outside minimum and maximum source range values were discarded.

3.3.5. Source apportionment of sediment sources

The relative contributions of the different sediment sources were determined by using different methodologies as shown in the different chapters.

In Chapter 5, it was used a frequentist multivariate mixing model proposed by Collins et al. (1997). The model solves a linear equations system, based in a conservative mass balance, multiplying different proportions of each of the sources and the sum between them. Results always reach a hypothetical sample that totals 100%. The procedure iterates a defined number of times until the options that are closest to the proportion of the target sample are determined.

Robustness of the solutions were assessed by a mean goodness of fit (*GOF*, modified from Motha et al. 2003):

$$GOF = 1 - \left\langle \frac{1}{n} \times \sum_{i=1}^{n} \left| b_i - \sum_{j=1}^{m} \left(a_{i,j} \cdot x_j \right) \right| / b_i \right\rangle$$

where b_i is the value of tracer property i ($i = 1$ to n) in the bed sediment sample, $a_{i,j}$ is the value of tracer property i in source type j ($j = 1$ to m), x_j is the unknown relative contribution of source type j to the bed sediment sample, m is the number of source types, and n is the number of tracer properties.

In Chapters 6 and 7 the MixSIAR Bayesian tracer mixing model framework (Stock et al., 2018), implemented by Stock and Semmens (2016) as an open-source R package, was used to estimate the relative contribution of each source to the suspended sediment samples and the artificial mixtures.

The fundamental mixing equation of a mixing model is:

$$b_i = \sum_{j=i}^{m} w_j \cdot a_{i,j}$$

where b_i is the tracer property i ($i = 1$ to n) measured in a suspended sediment sample, $a_{i,j}$ is the value of the tracer property i in each source sample j ($j = 1$ to m), w_j is the unknown relative contribution of each source j to the suspended sediment sample. MixSIAR accounts for variability in the source and mixture tracer data with the ability to incorporate covariance data to explain variability in the mixture proportions via fixed and random effects (Stock and Semmens, 2016; Stock et al., 2018). This is especially useful in this study because of the collinearity between colour parameters of the different chromaticity coordinates. Hence, a discriminant function was not used to select an optimum group of tracers, as weak tracers can only improve model representation. MixSIAR was formulated by using sediment type as a factor and an uninformative prior (Blake et al., 2018). The Markov Chain Monte Carlo parameters were set as very long: chain length = 1,000,000, burn = 700,000, thin = 300, chains = 3. Convergence of the models was evaluated by the Gelman-Rubin diagnosis.

Finally, the End Member Mixing Analysis (EMMA) approach (Christophersen and Hooper, 1992; Hooper, 2003) was applied in Chapter 7. Source categories were considered as end members with a fixed composition, conservative and distinguishable between them, while sediment samples were a mixture of these end members. We apply the diagnostic tools described by Hooper (2003). First, a bi-variable scatter plots were performed to identify if the tracers behave linearly conservative in the sediment samples. The tracers suggested linearly conservative mixing when had at least a linear trend of “$r^2 > 0.5$, p-value <0.01” with at least one of the other tracers used (James and Roulet, 2006). Then, a principal component analysis (PCA) was performed on the standardized values of the correlation matrix. The values were standardized by subtracting the average concentration of each tracer and dividing it by its standard deviation. Residuals were defined subtracting the original value from its orthogonal projection.

3.3.6. Experiment on conservative behaviour of sediment properties

In Chapter 8, twenty-seven representative samples of potential sediment sources were introduced within the channel of Es Fangar catchment during a whole year to investigate the conservative behaviour of sediment properties (i.e. total C, N, S, ^{137}Cs, ^{210}Pb$_{ex}$, As, Ba, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Zn, Na, Fe, Ca, K and Mg and colour properties)

Samples were introduced in 5*7 cm white polyamide bag with a 25 µm diameter mesh and were sealed using cable ties. Sealed samples were placed inside a piece of the same mesh and were sealed again with cable ties. The double layer of polyamide mesh allowed water to flow avoiding an excessive material loss.

Samples were placed near the gauging station that close Es Fangar creek catchment (Figure 3.3). In this point the channel has a cross-section with a rectangular broad-crested weir for low water stages (see dimensions in Figure 8.2E and F, Chapter 8). Samples were located 70 cm downstream from the wide-crested weir concrete structure. Eight iron bars (70 cm each) were nailed in to the channel bed (ca. 30 cm) and samples were fixed to the metal bars using cable ties. In addition, four samples, were introduced inside a time-integrated sediment sampler (Phillips et al., 2000) fixed at 5 cm from the channel bed (Figure 8.2C, Chapter 8). After that, eight time intervals were stablished to collect the samples throughout the year (i.e. 7, 30, 60, 90, 150, 210, 270, 365 days). Sample properties and analysis were exposed in section 3.3.2. laboratory treatment and analysis.

Coefficients of variation expressed in % (i.e. dividing the sample standard deviation by the mean and multiplying it by 100) were calculated for all soil properties considered. Thus, the relative dispersion of every soil parameter data set can be determined and compared with data sets belonging to different soil parameters.

Coefficient of variation (CV) was calculated to measure samples time (samples introduced within the channel in different time intervals) and spatial variability. Time CV was also divided in four time intervals. The time intervals considered were the following: initial submersion (i.e. 0- days), the period with constant flow (wet period, 7-90 days), the period without flow (dry period, 150-270 days) and the whole year. To identify the spatial variability of the different soil parameters within the catchment, it has been used data from forest (n= 6), crop (n= 5) and scrubland (n= 5) source samples from Chapter 7. In addition, a Pearson and spearman correlation coefficients were performed to observe correlations between the different soil properties and grain size (expressed as SSA; m^2 kg^{-1}), and between soil properties and total C content in percentage, as an approximation to organic matter content.

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8. In-channel alterations of soil properties used as tracers in sediment fingerprinting studies

Preliminary results

In-channel alterations of the most common soil properties used as tracers in sediment fingerprinting studies. **Paper in preparation**.

9. Discussion and conclusions

The preceding chapters of this thesis assessed the hydro-sedimentary dynamics in two Mediterranean catchments affected by Global Change processes, especially sediment transport and sediment source variability at different spatiotemporal scales. In this chapter, a brief synthesis is presented of the most relevant results of the thesis, which are then discussed as a whole with a view to presenting a comprehensive interpretation of (i) the assessment of the hydro-sedimentary dynamics in the study areas (i.e. Sa Font the la Vila and Es Fangar), (ii) the sediment source fingerprinting results, in terms of improving applicability and reducing uncertainties, and (iii) estimating the role of hydro-sedimentary monitoring combined with sediment fingerprinting as tools for catchment management. Thus, the final objective of the overall analysis of the thesis body will serve to refute or validate the two hypotheses presented in Chapter 1.

9.1. Sa Font de la Vila and Es Fangar hydro-sedimentary dynamics

Chapters 4, 5 and 6 focus on the fire-affected catchment Sa Font de la Vila (4.8 km^2). A nested catchment approach was used covering the 1.2 km^2 Sa Murtera sub-catchment, and water and sediment yields were measured at the outlet of both sites. Sediment origin was investigated using sediment fingerprinting and ^{137}Cs, ^{210}Pb$_{ex}$ and colour parameters as tracers.

During the first three post-fire hydrological years (i.e. 2013-14 to 2015-16) the average sediment yields were 1.6 and 6.3 t km^{-2} yr^{-1} for Sa Murtera and Sa Font de la Vila respectively. These values can be considered rather low in comparison with other burned catchments (see Table 4.1 in Chapter 4). However, they are higher than those obtained in other non-burned terraced catchments of the island of Mallorca (Estrany et al., 2009; Fortesa et al., 2021). Water yield in Sa Font the la Vila was also extremely low, despite the fact that the annual precipitation totals did not significantly deviate from the long-term average totals (see section 4.4.1. in Chapter 4). Counter-clockwise hysteretic loops between suspended sediment concentration and discharge dominated in Sa Font the la Vila (60%) during the study period. Normally, this kind of loop shape is ascribed to relatively distanced sediment sources (Oeurng et al., 2010; Williams, 1989), which could be fire-affected hillslopes in the study area. The percentage of counter-clockwise loops was higher during the first post-fire year (67%) coinciding with the highest annual sediment yield. Both values decreased over the following years. The percentage of counter-clockwise loops percentage can be considered high in comparison with other non-burned Mediterranean catchments (López-Tarazón and Estrany, 2017; Oeurng et al., 2010; Rovira and Batalla, 2006; Seeger et al., 2004), probably due to the wildfire perturbation. This hypothesis is reinforced by the results presented in Chapters 5 and 6. In the former, ^{137}Cs and ^{210}Pb$_{ex}$ were used as tracers to evaluate bed sediments' source ascription during the first post-fire flush in Sa Coma Freda creek (the east sub-catchment of Sa Font de la Vila catchment; see figure 5.2A). Results showed an average source contribution from burned hillslopes in the upstream part of 67%, reaching 75% in the downstream part (see the different parts of the stream in Figure 5.2B). In addition, in Chapter 6 we analysed suspended sediment origin during 4 floods which occurred between 2013 and 2015, including the first post-fire flush, in the Sa Murtera sub-catchment and the Sa Font de la Vila catchment. In general, tracing results using ^{137}Cs, ^{210}Pb$_{ex}$ and colour parameters showed a larger relative contribution from burned hillslopes during the first flood events. A contribution that gradually decreased with time (see Chapter 6). This results coincide with the observed decrease in the relative contribution of sediment originating in burned sources in Chapters 4 and 5.

As regards the prospect of increased erosion rates, runoff coefficients and sediment delivery in burned areas as documented by several authors (e.g. Shakesby and Doerr, 2006; Vieira et al., 2015), the results presented in Chapters 4, 5 and 6 appear to be reliable. However, other studies showed remarkable divergences in landscape response after a wildfire (Estrany et al., 2016; Owens et al., 2012; Smith et al., 2011; Wilkinson et al., 2009) (see section 6.5.2. of Chapter 6). Here it is hypothesized that the gradual decrease in sediment contribution from burned areas was not only due to a partial vegetation recovery, but also to the fact that the rainfall intensity thresholds generating effective slope-to-channel connectivity were not reached during the second post-fire hydrological year (Calvo-Cases et al., 2003; Li et al., 2019), resulting in partial sediment disconnexion of burned hillslopes.

In the Chapter 7 hydro-sedimentary continuous monitoring was combined with sediment fingerprinting (using ^{137}Cs, ^{210}Pb$_{ex}$ and colour parameters as tracers) to investigate suspended sediment origin in the Es Fangar catchment. Previous investigations determined that during the period 2012-2017 the annual runoff coefficient in the catchment ranged from 2.9% to 14.2% (average of 10.4%), whereas the quickflow contribution ranged from 9.9% to 45% (average of 33%), illustrating a huge inter-annual variability in the rainfall-runoff relationship (Fortesa et al., 2020b). During the same period, 80% of the sediment load was exported during autumn and winter, with an annual average sediment yield of 5.38 t km^{-2} y^{-1}. Flood events depicted a wide intra- and inter-annual variability and a marked seasonality with 85.3% of events occurring during the wet season. Sediment tracing results from the MixSIAR unmixing model showed that sediment mainly originated from crop fields all along the study period (Sections 7.4.4 and 7.5.1 of Chapter 7), with no seasonal patterns or major source changes regarding flood hydro-sedimentary characteristics. A similar low variability in sediment origin was observed in other Mediterranean catchments (e.g. Uber et al., 2019). Clockwise hysteresis loops predominated in the Es Fangar (occurring during 52.9% of the monitored events), coinciding with the results by Fortesa et al. (2020a), what suggested in-channel sediment remobilisation and erosion from near stream areas (i.e. crops). On the contrary, anti-clockwise or complex hysteresis patterns (47.1% of the monitored events) are sometimes related with the activation of different sediment sources (De Girolamo et al., 2015). Nevertheless, the sediment fingerprinting results did not provide information about different sediment origin regarding hysteretic type, suggesting that the analysis of hysteretic patterns might not always accurately provide information about sediment origin (Smith and Dragovich, 2009; Vercruysse et al., 2017). In the Es Fangar catchment, the lithology, land uses and the presence of agricultural terraces and dry stone walls could partially explain the low sediment source variability during the studied period (Calsamiglia et al., 2018; Estrany et al., 2010). Scrubland and forest areas are located in the catchment headwaters, where vegetation protects the soils of the steepest hillslopes, reducing runoff and suspended sediment generation. Moreover, carbonate materials characterized by low sediment availability and transmission losses (Calvo-Cases et al., 2003; Li et al., 2019) dominate in the upper parts of the catchment, whereas crop fields dominate in the valley bottom, which are completely exposed during certain periods of the year. Furthermore, a large part of the channel banks is constrained by dry stone walls, limiting channel bank sediment contributions. Therefore, the transference of significant amounts of sediment from the channel banks to the fluvial network would only occur when a dry stone wall collapses, which has been observed only rarely in the catchment.

Climatic, geological, topographical and land cover features regulate the sediment delivery in catchments (Walling, 1983). Mediterranean catchments are characterized by high inter- and intra-annual rainfall variability, heterogeneous lithology and highly human modified land uses and landscape topographic features (Calsamiglia et al., 2018; García-Ruiz and Lana-Renault, 2011; Yair, 1983; Zdruli, 2014). Mediterranean catchments show the highest sediment yields in Europe. Vanmaercke et al. (2011) reviewed suspended sediment concentrations across Europe and concluded that concentrations exceeded 40 t km^2 yr^{-1} in 85% of the revised data, and exceeded 200 t km^2 yr^{-1}in more than 50% of the values considered. In this context, Sa Font de la Vila and Es Fangar catchment average sediment yields, 6.3 t km^2 yr^{-1} and 4.5 t km^2 yr^{-1} respectively, can be considered as low. This can be explained, in part, given the (dis)connectivity concept within the sediment connectivity framework, as "*the degree to which any limiting factor constrains the efficiency of sediment transfer relationships*" (Fryirs, 2013; Fryirs et al., 2007). The (dis)connectivity framework defines a conceptual model of relationships between sediment linkages -categorized as lateral, longitudinal or vertical linkages- and blockages -barriers, buffers and blankets (cf. Fryirs, 2013). The effectivity between the different linkages is generally determined by its spatial configuration, or the presence of disrupting blockages. The increase of landscape connectivity depends on the breaching of blockages. Here, Fryirs et al. (2013) interpret the blockages as switches that can be activated depending on the magnitude of the driving forces, which have to exceed the thresholds established for every blockage before connecting different landscape compartments. In the Sa Font de la Vila and Es

Fangar catchments, agricultural terraces are considered to be blockages, that disconnect the different catchment compartments even when the window of disturbance tends to be more open (Prosser and Williams, 1998), i.e. after a wildfire perturbation. Agricultural terraces are water and sediment decoupling structures that dramatically increase the activation thresholds. Therefore, terrace status and its spatial configuration are key factors that control the (dis)connectivity. This could explain the low sediment yields and source stability in Sa Font the la Vila and Es Fangar catchments. However, these thresholds can also be exceeded. This happened on 29th October 2013 in Sa Font de la Vila, when a high intensity storm generated a suspended sediment yield of 17 t km^2 yr^{-1} in only 15 minutes, 92% of the sediment load measured during the first three post-fire hydrological years. Therefore, interactions between hydro-meteorological factors and human disturbances determine the sediment origin and catchment-scale sediment flux in highly modified landscapes (Fryirs, 2017; Poeppl et al., 2020). The results obtained are associated to human modifications through traditional soil conservation practices (Calsamiglia et al., 2018; Estrany et al., 2010). Thus, the Sa Font de la Vila and Es Fangar geomorphic systems depend to a certain extent on the maintenance and restoration of the human structures that configure it, especially soil conservation structures. Otherwise, a rapid change in its operation could eventually result in increased sediment yields, soil loss and degradation, promoting a sediment cascade effect (Burt and Allison, 2009).

9.2. Thesis contributions to the sediment fingerprinting framework

In a recent review of the state of the art of the sediment sources fingerprinting technique, Collins et al. (2020) highlighted some emerging topics and outstanding issues related to the sediment fingerprinting research and application. Some of the most relevant issues have been discussed in this thesis, such as (I) the application of sediment fingerprinting to wildfire impacted landscapes (Chapters 5 and 6), (II) the combination of sediment fingerprinting with other approaches for catchment management (Chapter 7) and (III) the tracer conservatism issue (Chapter 8).

I. Application of sediment fingerprinting to wildfire impacted landscapes

In Chapter 5, fallout radionuclides (^{137}Cs and ^{210}Pb$_{ex}$) were used as tracers in the eastern sub-catchment of the Sa Font de la Vila catchment (Sa Coma Freda) to determine the relative contributions of sediment sources in terms of spatial provenance (burned vs. unburned) and source type (soil surface vs. channel bank) in bed sediments. As regards this first approach, looking at the post-fire sediment source determination, results indicated that 67% of the upstream bed sediment contribution was generated from burned hillslopes, reaching 75% in the downstream part of the catchment due to the propagation of sediment delivered from burned areas (see figure 5.2B to observe the stream different parts).

In Chapter 6, the sediment origin in Sa Font de la Vila was further studied. Colour soil parameters measured with a spectrometer and a standard office scanner were used for the first time as tracers to effectively discriminate between burned and unburned sediment sources in the Sa Font de la Vila catchment and the Sa Murtera sub-catchment. The main advantage of colour parameters is that they can be measured quickly, are cheap and the method is non-destructive. Hence, they might be used to take quick decisions on post-fire management, as well as to evaluate the success of these measures since they make it possible to follow up. The efficiency of soil colour parameters was evaluated by using unmixing artificial laboratory mixtures when applying the MixSIAR Bayesian tracer mixing model framework (Stock et al., 2018). The results showed average absolute errors of 12.3% ± 9.1, 12.3% ± 4.2 and 10.1% ± 4.2 for 2-, 3- and 4-source mixtures, respectively. Errors were of the same order of magnitude as errors obtained by other authors using other types of tracers (Brosinsky et al., 2014; Gaspar et al., 2019; Haddadchi et al., 2014). However, when the colour signatures were similar (e.g. mix 4-m4, mix4-m5, Supplementary Table 6.5), colour tracer measurements are not reliable enough to quantify source contributions. The changes in the soil's visible reflectance (Lentile et al., 2006) and its carbon content (Bodí et al., 2014) caused by the presence of ash, triggered the change of the upper soil layer colour. As a result, colour parameters estimated from diffuse reflectance laboratory measurements were able to discriminate between burned surface soil, unburned surface soil and channel bank sources. Artificial mixtures showed that most colour parameters were linear additive and, individually, were able to predict the colour of the mixtures by using a mass balance approach and, again, the errors were comparable with values reported in other studies (Gaspar et al., 2019; Martínez-Carreras et al., 2010b; Uber et al., 2019). Chromatic parameters calculated from the spectrometer in the laboratory and scanner-based colour parameters correlated closely (p < 0.01), confirming that colour parameters obtained with an office scanner can be as reliable as colour tracers from a spectrophotometer. Scanner measured colour parameters were used in other fingerprinting research by Pulley et al. (2016). They compared colour and mineral magnetic signatures to trace bed and suspended sediment in the South African Karoo. The discriminatory efficiency of colour signatures ranged between 92.2% and 96.7% and were comparable to the results obtained using mineral magnetic signatures (i.e. 94%). In the Sa Font de la Vila catchment, colour parameters were compared with ^{137}Cs and ^{210}Pb$_{ex}$ as tracers. Estimated source ascriptions with both methods matched when it came to predicting the dominant source in 7 of the 9 samples measured. Thus, colour tracers proved to be useful in discriminating between burned and unburned sources. Therefore, it can be considered suitable for suspended sediment source ascription and monitoring as part of post-fire management strategies.

After a wildfire, the hydro-sedimentary response in affected landscapes can be drastically altered (Moody et al., 2013; Shakesby, 2011; Shakesby and Doerr, 2006). In general, surface runoff and sediment yield from hillslopes is increased (Candela et al., 2005; Scott et al., 1998), promoting erosion and a higher slope-to-channel sediment transfer that may generate downstream impacts related to fine sediment transport and its associated pollutants (Collins et al., 2017). Wildfires are one of the disturbances that can be increased as a result of global change processes (Huang et al., 2014) such as increased temperatures, variations in rainfall patterns and land use changes. The potential risks associated with wildfires can seriously impact the ecological status of the environment, and therefore are attracting increasing interest from researchers and landscape managers (Robinne et al., 2020; Smith et al., 2011; Wohl, 2018). Chapters 5 and 6 are focused on research into the effects of wildfires on soil properties and hydrological and geomorphological processes by using the sediment fingerprinting technique. There are not many studies that discriminate between burned and unburned sources (e.g. Estrany et al., 2016), so the contributions in these chapters are valuable in their own right. However, the greater novelty lies in the development of the use of colour coefficients to investigate changes in soil properties (i.e. incorporation of ash) and sediment sources following a wildfire. These colour fingerprints, in combination with fallout radionuclides, were used to understand geomorphological processes following the 2013 wildfire which occurred in the Sa Font de la Vila catchment. The results obtained may be relevant for future studies that use sediment fingerprinting to understand how catchments respond to natural and anthropogenic disturbances, especially wildfires. However, further investigation needs to be done on the application of sediment fingerprinting in fire affected landscapes, particularly in relation to the differential effects of fire on sediment properties (Collins et al., 2020) and its evolution over time.

II. Combination of sediment fingerprinting with other approaches

In Chapter 7, the hydro-sedimentary dynamics in Es Fangar Creek were linked with the uses of soil colour parameters and fallout radionuclides as tracers within an integrated approach to predict dominant suspended sediment sources. A Bayesian mixing model (MixSIAR; Stock et al., 2018) and an End Member Mixing Analysis (EMMA; Christophersen and Hooper, 1992) were applied. The selection of tracers was problematic. Fallout radionuclides did not discriminate between channel bank and crop soils, and colour parameters did not discriminate between forest and scrub sources. In addition, even though colour parameters discriminated between channel bank and crops their spectral signatures were relatively close (Figure 7.4).

To combine fallout radionuclides and colour parameters in the same tracer set, sources were grouped into only two categories (i.e. channel-crop and forest-scrub) in a first un-mix to accommodate a larger number of tracers. The unmixing process was repeated using only colour parameters so as to be able to discriminate between three sources (i.e. channel bank, crop soil and forest-scrubland as a single source). MixSIAR identified channel bank-crops soil as the dominant sources in the two source analysis and crop soils in the three source analysis. In addition, comparing MixSIAR results between the two and three sources analysis, the mean absolute error in the source apportion percentage prediction was relatively low (1.2%) (i.e. adding up crop and channel bank % results for the three source analysis), which shows that the model is robust using the selected tracers set in both analyses. EMMA showed similar results. The source tracer values plotted in the U1–U2 mixing diagram of suspended sediment tracers showed that it was possible to observe that sediment samples were clustered close to the crop and channel bank signatures (Figure 7.10). Hence, results revealed that the approach is able to determine the main sediment sources. Furthermore, as in the MixSIAR results, sample 2 was plotted closer to the forest and channel bank signatures. Our analysis suggests that the EMMA approach can be a good choice when it comes to identifying dominant sources of sediment using simplified procedures, in comparison with standard mixing models.

In this Chapter 7, it was found that EMMA, a statistically simpler model compared to MixSIAR and rarely used in sediment fingerprinting (Munkundan et al., 2010; Rose et al., 2018), can robustly identify the main sediment sources. Therefore, its use may be appropriate in sediment fingerprinting analysis alone or as a complementary model to check the robustness of results obtained with other models. However, the main purpose was to combine hydro-sedimentary monitoring with sediment fingerprinting to determine the main factors regulating sediment source contributions. Although floods were grouped into four clusters based on its hydro-sedimentary characteristics and hysteretic loops analysis, it was not possible to establish a correlation between sediment origin and hydro-sedimentary variables, because sediment sources showed a low variability in the Es Fangar catchment during the study period.

Few studies combined suspended sediment source assessment with the analysis of hydro-sedimentary response at the catchments scale. Navratil et al. (2012) combined river/rainfall monitoring and sediment fingerprinting using fallout radionuclides and geochemistry in a 905 km^2 catchment located in the French Alps. They observed how the ca. 80% sediment load occurred during widespread and long rainfall events with low intensities, while shorter storms were associated with higher discharge peaks and suspended sediment concentrations. However, and despite the high intra- and inter-flood variability of the analysed flood sediment sources remained relatively stable. On the contrary, Vercruysse and Grabowski (2019) found interesting source variations regarding hydro-meteorological drivers in an 879 km^2 catchment in UK. On the one hand, street dust and limestone grassland sources were strongly correlated with suspended sediment concentration, discharge and 1-day antecedent rainfall. On the other hand, millstone and coals grassland sources were mainly correlated to antecedent hydro-meteorological conditions (e.g. precipitation and discharge).

In the Es Fangar catchment, the association between fingerprinting and sediment monitoring did not show any pattern because the agroforestry terraced landscape mosaic over carbonate rocks combined with the lack of extreme flood events during the study period, avoided the activation of remote or inaccessible sediment sources during frequent floods. However, the combination of these different approaches could provide information on the activation thresholds for each of the sediment sources considered. A good knowledge of the relationships between hydro-sedimentary dynamics and the activation of different sediment sources can help managers to define optimal intervention strategies, but can also be used to create optimized quantitative (in terms of amount) and qualitative (in terms of origin) sediment models to be integrated in catchment management plans. Thus, further research is necessary to reach a comprehensive understanding about erosion and sediment transport dynamics in the Es Fangar catchment.

III. Tracer conservatism

Finally, in Chapter 8 an experiment was performed to investigate eventual in-channel changes occurring to the most common soil properties used as tracers in sediment fingerprinting studies. Preliminary results in the Es Fangar Creek catchment showed a low variability in sediment properties during the study period with an average in-channel coefficient of variation of 8.1 ± 8.8% for all properties in different land uses (i.e. forest, crop and scrubland). The average spatial variability of sediment properties (16.3 ± 18.5) was higher in comparison with in-channel variation in the 90% of the comparable observations (i.e. all parameters for every land use except S, green, blue, cie X, cie Y, cie Z and cie L in scrubland samples). Colour properties were the most stable tracers with in-channel average CVs for 365 days of 2.6 ± 2.2%. This low variability in colour parameters was similar to that in other studies of shorter duration (Legout et al., 2013; Poulenard et al., 2012; Uber et al., 2019), confirming that colour parameters can be used as stable tracers (i.e. conservative) in sediment fingerprinting. Geochemical elements showed a more heterogeneous behaviour. The more stable elements during the study period were Ba, Cd, Co, Cr, Cu, Mn, Ni, Pb, Zn, Fe, Ca, K and Mg with in-channel average CVs for all land uses in 365 days ranging from 5.4 to 16.9%. On the contrary, As, Mo and Na presented higher variability (CVs 22.6 to 32.2%). C, N and S showed less in-channel variability (average in channel CVs for all land uses in 365 days of 2.1 ± 0.4% and 8.1 ± 6% and 26.8 ± 14.9% for C, N and S respectively) in comparison with its spatial variability (average CV of 28.8 ± 12.8% and 37.9 ± 19.7% and 35.5 ± 21.1 respectively for all three land uses). Finally, fallout radionuclides in-channel average CVs for all land uses were 19.4 ± 8.7 for ^{137}Cs and 25.7 ± 7.5 for ^{210}Pb$_{ex}$. In general, the three land use types showed similar variability in soil properties, being slightly higher in crop samples. Comparing crop and TIS samples (i.e. samples within time integrated suspended sediment traps; average in-channel CV for all properties for 365 days of 9.3 ± 0.1% and 8.6 ± 9.2% respectively), it can be stated that lack of direct insolation and likely differentiated humidity and temperature conditions did not have significant effects on soil properties. The general low variability observed in soil properties, and its strongest correlations with SSA and C further emphasizes the role of particle size and organic matter in the conservative behaviour of soil properties (Koiter et al., 2018).

An increase in our knowledge of tracer conservativeness is one of the main issues as regards sediment fingerprinting (Collins et al., 2020; Koiter et al., 2013; Lizaga et al., 2020). To use a soil property as a tracer it should be representative of main erosion sources, it must be able to differentiate between them, must be measurable and remain stable or vary in a predictable way over time and space (Motha et al., 2002). However, some alteration processes are known to occur during mobilization and mixing along hydrologic pathways. The degree of alteration depends on the stability of the marker and is highly site dependent. Therefore, this is difficult to address and is not often considered beyond defining the set of tracers with conservative behaviour by performing statistical analysis (e.g. Collins and Walling, 2002; Collins et al., 1997; Smith et al., 2018; Walden et al., 1997; Walling, 2005; Wilkinson et al., 2013).

Few studies have investigated the conservative behaviour of soil properties (Koiter et al., 2018; Legout et al., 2013; Motha et al., 2002; Poulenard et al., 2012; Uber et al., 2019). In Chapter 8 an investigation was conducted into eventual in-channel changes occurring to the most common soil properties used as tracers in sediment fingerprinting studies. The novelty relies (I) in the characteristics of the study area (i.e. Es Fangar), a Mediterranean catchment with an intermittent flow regime with marked dry and wet seasons and (II) the duration of the experiment exposing the sediment over large time periods (i.e. one year) with different hydro-meteorological conditions. In the experiment, soil samples were exposed to natural variables (e.g. temperature, pH, moisture), covering the intra-annual hydro-meteorological variability in Mediterranean environments. Therefore, the conditions can be considered optimal when it comes to assessing possible changes over time in Mediterranean catchments or temporary rivers.

Despite its limitations (Chapter 8, section 8.5.2), such a study under this time scale may be useful for evaluating the application of certain tracers in fingerprinting studies, especially in Mediterranean environments where hydro-meteorological conditions exhibit a great contrast throughout the year. However, it is necessary to continue iterating as regards the study of the tracers' conservativeness, considering the possible transformations which occurred in hillslopes and channels, so as to reduce the uncertainties in sediment fingerprinting.

9.3. Sediment fingerprinting and hydro-sedimentary monitoring as tools for catchment management in Mediterranean environments

One of the most relevant aspects of scientific knowledge for society is its applicability. In this regard, applied geomorphology could be defined as the "branch of science within the broader discipline of geomorphology that focuses on geomorphological landforms and processes of societal concern" (Meitzen, 2017). Therefore, it is the usefulness of geomorphological scientific knowledge when it comes to solving socially relevant problems by helping, among others, landscape managers with decision-making on numerous issues of social relevance. The general objective of the present thesis was "*To identify erosion and sediment transport processes (functional connectivity) in two Mediterranean catchments affected by different global change processes at different spatio-temporal scales, by improving current techniques for* *sediment origin determination (i.e., reducing uncertainties, time and cost) for its better implementation in catchment management plans*". Therefore, apart from a better hydro-sedimentary dynamic understanding of the study areas and the optimization of the sediment fingerprinting technique, results had to be useful for catchment management.

Integrated catchment management plans need to reach a comprehensive understanding about erosion and sediment transport dynamics to effectively develop integrated management approaches (McCarney-Castle et al., 2017; Owens et al., 2005). Therefore, one needs to assess the sediment cascade between upstream erosive processes, sediment mobilization through hillslopes and within the channel, and downstream sediment yields to mitigate its possible negative effects. Sediment fingerprinting and river hydro-sedimentary monitoring and analysis were useful approaches to monitoring and controlling sources of soil and sediment erosion for a variety of different land uses and environments. Here it is argued that the combination of sediment fingerprinting and river hydro-sedimentary monitoring improves our knowledge about all these processes and, therefore, are essential tools in management plans.

Sediment source fingerprinting has been widely used in recent decades to detect fine sediment sources at catchment scale (Collins et al., 2020; Davis and Fox, 2009; Guzmán et al., 2013). Interesting methodological guides have been published aimed at end-users as a basis for correctly applying the technique (Collins et al., 2017). During the development of this thesis, the use of economic tracers –in terms of time and economic cost– has been emphasized to simplify the application process of sediment fingerprinting technique. As in previous studies (e.g. Barthod et al., 2015; Martínez-Carreras et al., 2010a), colour parameters have been shown to have a comparable efficacy in contrast with more established tracers (i.e. ^{137}Cs and ^{210}Pb$_{ex}$), even in wildfire affected landscapes (Chapter 6). In addition, there is the possibility of performing the measurements using a standard office scanner with good results (Pulley and Rowntree, 2016; Chapter 6) obtaining, on this occasion, correlations of $p<0.01$ with the measurements made with a laboratory spectroradiometer. Sediment tracing by using colour parameters allows rapid samplings and pre-treatments, because of the low mass required to perform the measurements (ca. 1-2 g) and the possibility of using an office scanner to measure the samples. In addition, the probably low in-channel variability of colour tracers detected in comparison with the rest of the tracers evaluated in Chapter 8, highlighted colour parameters as a remarkable tracer for use in catchment management plans. Another interesting point investigated during the development of the thesis was the use of the EMMA approach, which is not based on a mixing model but on a principal component analysis of the data set (PCA). It is argued that a quantitative estimation of the sediment apportion of the different suspended sediment sources in a catchment might not always be needed when implementing sediment management plans, but rather an accurate identification of dominant sources. EMMA offers advantages based on simplified procedures in comparison with the more extended mixing models, what could result in easier implementation.

Finally, the combination of sediment fingerprinting and continuous hydro-sedimentary monitoring was proposed, to assess the factors that control suspended sediment transport as a surrogate of erosion problems in river catchments. Despite being complementary, the integration of these approaches to detect activation patterns for different sediment sources or disconnected catchment compartments is not usual (Evrard et al., 2011; Navratil et al., 2012; Vercruysse and Grabowski, 2019). A detailed study about hydro-sedimentary characteristics of flood events, linked to the spatiotemporal variation of suspended sediment sources can allow the detection of the predominant sediment sources based on the flood hydro-sedimentary dynamics, as well as the (dis)connectivity thresholds that can activate it. The acquisition of this knowledge could be relevant to develop sediment transport models that integrate information on sediment origin (Owens et al., 2005; Perks et al., 2017; Vercruysse et al., 2019), and useful for detecting drastic changes in catchment geomorphological processes as well as predicting Global Change impacts.

In Chapter 7, sediment fingerprinting and hydro-sedimentary monitoring were combined to determine the main factors regulating sediment source contributions, and evaluate the potential of hydro-sedimentary monitoring combined with sediment fingerprinting as a sediment management tool in Mediterranean catchments. The stability of the Es Fangar catchment in terms of sediment source contributions made it impossible to find correlations between sediment origin and hydro-sedimentary variables. However, in other study areas it has been possible to establish correlation patterns (Vercruysse and Grabowski, 2019). Throughout, it is desirable to continue research into this issue, especially in highly variable environments such as the Mediterranean basin.

9.4. Limitations and future perspectives

Research in geomorphology encompasses a series of difficulties, which can sometimes turn into limitations. In this type of science, the laboratory is the field, so there are numerous factors that can interact, varying the initial planning of the research proposal. The methodological development of any experiment in catchment geomorphology involves considerable effort in an environment that cannot be controlled.

The main limitations of the thesis are listed below, as well as some proposals for future work:

Chapter 4: One of the most evident limitations was the lack of data for the first post-fire year in the Sa Murtera sub-catchment caused by technical problems with the turbidity probe into the gauging station. This problem did not make it possible to monitor the hydro-sedimentary processes in a part of the Sa Font de la Vila headwaters where the hillslope-channel coupling is higher and when the landscape was more vulnerable to erosion. In addition, a multivariate correlation analysis, a principal component analysis to group variables and the application of a quantitative hysteresis index (e.g. Zuecco et al., 2016) could have been carried out to achieve a better understanding of the hydro-sedimentary response of Sa Font de la Vila. Opportunely, the continuous water and sediment monitoring has continued over time. This will allow an exhaustive analysis of the evolution of a Mediterranean catchment after a fire for more than 7 years. These data, in conjunction with sediment origin analysis (i.e. sediment fingerprinting), could provide valuable information during the medium and long-term for assessing geomorphological processes in fire-affected catchments.

Chapter 5: The limitations in this chapter are mainly due to the lack of a larger set of tracers, the lack of artificial samples to check the reliability of the tracers when predicting source proportions in sediment samples, and not applying a complementary mixing model to contrast the results and the reduced number of samples from unburned sources compared to burned sources. However, all these limitations have been solved in later works as reflected in Chapter 6.

Chapter 6: The main limitations were the absence of an annual source resampling, to check tracer evolution and the use of ash from another catchment for an experiment on its influence on the colour tracers. However, these limitations were solved in part by using soil samples collected in a headwater micro-catchment in 2016 and by using the above-mentioned ash samples collected in a catchment with similar characteristics. Future work can involve the use of novel tracers (e.g. compound specific stable isotopes, biomarkers) in other sediment fingerprinting research, the combination of hydro-sedimentary data with sediment origin to identify the activation patterns for different sediment sources, the use of a different source categorization (e.g. land uses) or even by performing a tracer conservativeness experiment as in Chapter 8.

Chapter 7: It was not possible to establish patterns between flood event hydro-sedimentary characteristics and sediment origin. Here a larger set of tracers might have helped to discriminate more robustly between source types. In addition, a resampling of the sources could have been proposed or a larger number of samples could have been included for each type of land use. Future work can be focused on solving the previously explained limitations by carrying out a source resampling or by including novel tracers in the sediment origin analysis. In addition, the continuous monitoring of water and sediment fluxes in the gauge station installed in Es Fangar, will make it possible to repeat the study and, perhaps, observe the activation of the different sediment sources in Es Fangar and link it with flood characteristics.

Chapter 8: The limitations were associated with the difficulty of conducting the experiment in natural conditions, as well as the large amount of time and resources that is required. The main limitations were the inability to integrate the physical effects on sediment particles derived from the transport in suspension (e.g. abrasion, aggregation), the possible influence of nylon bags and the steel bars on some soil properties, the absence of samples sieved to different particle sizes or the lack of different replicas for each sample. In addition, in-channel transformations only represent a part of the potential property changes during the erosion and sediment delivery processes. Therefore, in the evaluation of the conservative behaviour of a tracer, the processes generated on hillslopes should also be considered (Koiter et al., 2018; Motha et al., 2002). Here, for future works on tracer conservativeness we will work with 15 artificial source mixtures with different known proportions generated and submerged during the experiment. The possibility of performing an analysis of the proportions of the artificial mixtures by using “sources” such as the original soil material exposed to different in-channel intervals with different hydrological conditions, will verify the reliability of the variability/stability results for all the tracers analysed.

9.5. Conclusions

The hydro-sedimentary dynamics and suspended sediment origin has been investigated in two Mediterranean catchments affected by different Global Change processes. Despite the limitations explained in section 9.4, the combination of sediment fingerprinting with continuous hydro-sedimentary monitoring made it possible to assess its hydro-sedimentary dynamics during the study period. In Sa Font de la Vila, results showed a gradual decrease from burned source contributions over time, while in Es Fangar the contributions from crops sources dominated throughout the study period without substantial changes. Sediment yields were 6.3 t km^{-2} yr^{-1} and 4.5 t km^{-2} yr^{-1} for Sa Font de la Vila and Es Fangar respectively, being low in comparison with other Mediterranean catchments. This was mainly attributed to catchment lithology, land uses (in Es Fangar), vegetation recovery (in Sa Font de la Vila) and the presence of agricultural terraces and dry stone walls.

The use of soil colour parameters as tracers has been successfully proven in the two catchments, confirming its suitability for use in sediment fingerprinting as a rapid and economic tracer, even in fire-affected catchments. In addition, the strong correlations between the measures made with a laboratory spectroradiometer and a standard office scanner make colour parameters even easier to use in research and management plans. Moreover, the long duration tracer conservativeness experiment which was carried out, despite its limitations, confirmed that in-channel changes suffered by all the analysed tracers were generally lower than their own spatial variability within the catchment. Additionally, colour parameters were the most stable tracers. These statements contribute to the improvement of the sediment fingerprinting technique, so it can be said that **hypothesis 1**: *"optimization of the sediment fingerprinting technique through research on the conservative behaviour of soil parameters and the use of low-cost and fast-to-measure tracers allowing evaluation of some of the assumptions underlying the technique, improvement of its applicability and the reduction of uncertainties"* is **confirmed**.

It was not possible to identify the activation patterns of different sediment sources by using a combination of hydro-sedimentary monitoring and sediment fingerprinting. This was caused mainly due to the Es Fangar catchment stability in terms of suspended sediment origin (mainly from crop sources). Like the low sediment yields recorded, the sediment source stability is attributed to lithological characteristics, land uses and the presence of agricultural terraces in the study area. However, events of higher magnitude could exceed the sedimentary (dis)connectivity thresholds of the rest of the sources considered, and activate them. Therefore, and despite the fact that in other similar studies it has been possible to establish links between the hydro-sedimentary behaviour and the origin of the sediment, in this case **hypothesis 2**: "*hydro-sedimentary monitoring combined with sediment fingerprinting makes possible a better identification of the activation patterns of the different sediment sources within a catchment, resulting in a useful tool for catchment management*" is **partially refuted**. The links between flood events characteristics and sediment origin could not be established. However, it has been proven that hydro-sedimentary monitoring and sediment fingerprinting are useful tools for integrated catchment management plans in Mediterranean environments.

9.6. References

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