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Combining catchment modelling and sediment fingerprinting to assess sediment dynamics in a Spanish Pyrenean river system.

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Abstract:

The elevated supply and transfer of sediment in river systems can impact on water resource infrastructure, water quality and ecosystem health around the world. Moreover, the loss of reservoir capacity due to siltation is of particular concern in regions that experience water scarcity, such as the Mediterranean. Therefore, quantifying the sediment production from contributing catchments to reservoirs is of major interest to support the development of management plans for maintaining reservoir sustainability. Within this context, sediment production from a catchment in the central Spanish Pyrenees was investigated by combining the outputs of the Soil and Water Assessment Tool (SWAT) model with sediment source fingerprinting. The study examined the large alpine-prealpine catchment of the Barasona reservoir, which is an agroforest catchment supplying sediment to the reservoir at an annual rate of around 3.5 t ha^{-1} . For the period 2003-2005 the simulated specific sediment yields for the catchment by SWAT model are mostly below $2 \text{ t ha}^{-1} \text{ year}^{-1}$ with maximum values reaching $250 \text{ t ha}^{-1} \text{ year}^{-1}$. Simulated sediment production is dependent on (i) land use/cover, (ii) precipitation amounts and (iii) slope gradients. The highest sediment yield was simulated for the badlands, in the wettest year and for the greatest slope gradients. The predicted catchment source contributions obtained for the reservoir sediments were low ($<7\%$) for forest and scrubland sources, greater for agricultural sources and exceeded 50% for subsoil sources. Results from both procedures indicate that badlands are the main sediment source while agricultural land is of secondary importance. This combination of procedures allowed catchment sediment production and source contributions to be linked to reservoir infilling. It also demonstrates the benefits of combined modelling

and sediment fingerprinting approaches in complex catchments where siltation threatens water and energy security.

Keywords: SWAT model, source production, mixing model, source ascription, large mountain catchments

Abbreviations: SWAT: Soil and Water Assessment Tool; HRUs: Hydrological Response Units; GOF: goodness of fit; RMSE: root mean squared error; KW: Kruskal–Wallis *H*-test; DFA: discriminant function analysis; SSY: specific sediment yield.

1. Introduction

Sediment has a variety of roles and impacts and its regulation and management are complex and scale dependant (Cerdá et al., 2013). A recent review of European sediment yields demonstrated that Mediterranean rivers have higher yields than those in the rest of Europe, which has been attributed to climate, topography, lithology and land use (Vanmaercke et al., 2011). In this context, reservoir siltation represents a critical off-site problem derived from soil erosion and sediment delivery within the Mediterranean environment (Navas et al., 2004), and its effects can be economically and societally serious in terms of both water and energy security. This fact is particularly important for those in mountainous areas, such as the Pyrenean region where high erosion rates rapidly reduce reservoir storage capacity (Navas et al., 2008). Moreover, human activity during the last 4,000 years has contributed to the disturbance of the original landscape and its hydromorphological dynamics (Navas et al., 1997; García-Ruiz and Valero-Garcés, 1998), triggering in most cases siltation problems (Valero-Garcés et al., 1999; Navas et al., 2009). Changes in both hydrological regimes and the supply of sediment loads have been recorded in Pyrenean dammed rivers and lakes (Morellón et al., 2011; Navas et al., 2011).

In mountainous environments, problems associated with sedimentation are exacerbated by the fact that the bulk of sediment is exported within very short periods, e.g. after high magnitude storms or during the annual snowmelt (Meybeck et al. 2003). Sediment supply from mountainous catchments to lowland rivers is largely controlled by local erosion processes and the availability of sediments from highly erodible materials (e.g. Mathys et al., 2005; Schuerch et al., 2006; Nadal-Romero et al., 2008; Wang et al., 2009; Skalak and Pizzuto, 2010; Evrard et al. 2011). Hence, information on the risk of water-induced soil erosion and sediment export in mountain areas is essential in order to implement appropriate and effective control measures along the downstream river network.

Direct continuous measurements with sufficient spatial coverage to develop knowledge of catchment sediment yield are difficult to obtain in large river systems, especially if these catchments are in mountainous regions. Consequently, workers have increasingly turned to indirect measurement tools to examine sediment fluxes. Catchment sediment production and yield modelling has already been proven effective when used in combination with sediment source fingerprinting procedures (e.g. Theuring et al., 2013, Wilkinson et al., 2013; Palazón et al., 2014). The catchment models (e.g. SedNet or SWAT) allow assessment of differing inputs from various source types and their spatial origin within the catchment, whereas the fingerprinting procedures allow identification of the proportional sediment source contributions to the mixtures of sediment derived from contributing catchments.

The Soil and Water Assessment Tool (SWAT) has been extensively applied to deal with a wide range of scales and issues related to hydrology, water management, climate change impacts, land use impacts, best management practices, sedimentation, and pollution (Gassman et al., 2007). SWAT has been widely used for hydrological

simulations in mountainous catchments (e.g. Zhang et al., 2008; Narula and Gosain, 2013). However, there have been few studies that evaluate sediment production in alpine mountain catchments (e.g. Abbaspour et al., 2007; Flynn and Van Liew, 2011; Gamvroudis et al., 2015) and very few studies have been conducted to quantify sediment production from land uses in large alpine–prealpine catchments (Palazón and Navas, 2016).

Sediment source fingerprinting procedures have been increasingly used to assess the proportional contribution of sediment sources to target or mixture samples (e.g. Collins et al., 2010a, b; Martínez-Carreras et al., 2010b; Blake et al., 2012; Schuller et al., 2013; Tiecher et al., 2015). A review of the procedures and applications of sediment source fingerprinting techniques can be found in Walling (2013). These techniques are based on the identification of differences in the tracer properties or ‘fingerprints’ of the potential sediment sources on the basis of statistical analysis and interpretation. Together with the increasing use of this procedure, there has been a growing focus on the accuracy of predicted source contributions (e.g. Haddadchi et al., 2014; Laceby and Olley, 2015). Recently, Palazón et al. (2015b) proposed an approach for assessing the accuracy of the sediment fingerprinting procedure by using virtual mixture samples and the root mean squared error (RMSE).

Combination of modelling and tracer-based techniques at the large catchment scale is likely to enhance the knowledge about erosion processes, thereby increasing the robustness of the simulated results. In this context, the importance of the Barasona reservoir, located in the Spanish Pyrenees, as a source of water for irrigation to the lowlands and its siltation management problems have been investigated since the 1990s (e.g. Avendaño-Salas et al., 1997; Navas et al., 1998; Valero-Garcés et al., 1999). From these studies, those which identified sediment sources pointed to the badlands

developed in Eocene marls as the main source of sediment (e.g. López-Tarazón et al., 2015). Some of them also identified agricultural lands as the secondary source of sediments (e.g. Alatorre et al., 2010; Palazón and Navas, 2014, 2016). These previous research findings of the Barasona catchment together with the successful combination of indirect techniques (SWAT and sediment fingerprinting) applied to the smaller headwater catchment to assess surface contributions from soil types to a small reservoir (Palazón et al., 2014) provided the basis for exploration of the approach at the wider catchment scale as reported in this contribution. To develop knowledge of catchment-wide erosion processes that contribute to reservoir siltation, the outputs from the application of SWAT model, calibrated and validated in Palazón and Navas (2014), were combined with the source apportionment data obtained using the sediment fingerprinting technique. In this context, the main objectives of this study are: (i) to assess and quantify the spatial distribution of sediment production from the range of land use/covers in the Barasona catchment using the SWAT model and compare outputs with sediment source apportionment results derived for reservoir sediment mixtures using sediment fingerprinting and (ii) to assess the accuracy of the sediment fingerprinting procedure at the larger catchment scale by conducting an auto-evaluation with virtual mixture samples.

2. Material and methods

2.1 Study area

The Barasona reservoir, located in the central part of the Spanish Pyrenees, collects the discharge of most of the Ésera River catchment and, its main tributary, the Isábena River catchment, integrating an effective drainage catchment of 1479 km². Part of its headwater (30 km²) is disconnected to the effective drainage catchment as it drains through a karstic system (Fig 1; Palazón and Navas, 2013). The hydrologic regime of

the catchment is transitional nivo-pluvial, characterized by two maxima (García-Ruiz et al., 2001). The Ésera River is regulated by small reservoirs, canals and dams, whereas, the Isábena River is non-regulated. In general, the main rivers flow through blocky or rocky channels.

The drainage catchment of the Barasona reservoir is characterized by an abrupt topography with an altitude range of about 3000 m, a mean elevation of 1313 m and an average catchment slope of 39% (Fig. 1). The lithological competence of the five main Pyrenean structural units (WNW–ESE-trending geologic units, Table 1 and Fig. 1a), within the catchment controls the distribution of the geomorphological processes and slope ranges (Palazón and Navas, 2014). Despite the fact that badlands on the Eocene marls represent less than 1% of the total catchment area (see location in Fig. 1b), they constitute the most important sediment source in the catchment (e.g. Alatorre et al., 2010).

Table 1.- Main characteristics of the Pyrenean structural units.

	Structural Units	Characteristics
North	Axial Pyrenees	- quartzites, limestone, shales, granites and granodiorites, -large mountain bodies and the highest Pyrenean peak (Aneto Peak 3404 m a.s.l.)
	Internal ranges	- large packages of limestones interbedded with marls and sandstones -deep and narrow gorges
	Internal depressions	- more erodible materials which develop badlands on marls - comprising depressions located into the previous structural unit
	Intermediate depression	- detrital sedimentary rocks - relative lowland area

South	External ranges	- sandstones and limestones - ranges that delimit the catchment to the south
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The catchment has a mountain climate, wet and cold, influenced by the Atlantic Ocean and the Mediterranean Sea (García-Ruiz et al., 2001). The conjunction of these influences together with the relief generates gradients in temperature and precipitation as recorded for both north–south and west–east regions. Annual precipitation and temperature range from 500 mm and 12 °C at the outlet (424 m a.s.l.) to more than 2500 mm and less than 4 °C on the highest divides (> 3000 m a.s.l.).

Climatic and topographic gradients from south to north also influence the distribution of the predominant land uses which varied from mostly cultivated land in the lowland southern areas, followed by forest in the intermediate ranges, to alpine grassland in the highlands. Forest and pastures occupy more than 50% of the catchment, followed by scrublands with more than 20% and cultivated land occupies around 15% (Fig. 1b). Important changes in land use occurred during the last 60 years in the Spanish Pyrenean region, resulting in substantial land abandonment that has affected most parts of the agricultural areas, triggering the subsequent process of natural reforestation (Navas et al., 2008).

In general, the soils of the catchment are young, stony and mostly shallow overlying fractured bedrock with textures ranging from loam to sandy loam. Soils are alkaline and generally well-drained with limited average water content and moderate to low structural stability.

Fig. 1 a) Location of the Barasona catchment in the Iberian Peninsula, the distribution of the rain and temperature gauge stations, the location of the karst system at the headwater, the distribution of the Pyrenean structural units and the distribution of the source samples for the Barasona catchment and the mixture samples in the Barasona reservoir; b) distribution of the soil types (FAO 2007), land uses and land covers and the DEM of the catchment and c) photos of the main land use/land cover sources and the Barasona reservoir filling.

The Barasona reservoir (692 ha) has suffered siltation problems since its construction in 1932 (Navas et al., 1998; Valero-Garcés et al., 1999). Its initial water capacity was 71 hm³, which was increased to a maximum storage capacity of 92 hm³ in 1972 and with maintenance operations in the 1990s (Alatorre et al., 2010) the reservoir has a current capacity of 85 hm³. A bathymetric survey in 1995 indicated that over its initial 65 years of operation the reservoir has lost one third of its original capacity. This initial bathymetric survey yielded a specific sediment yield of 3.50 t ha⁻¹ year⁻¹ (Avendaño-Salas et al., 1997).

2.2 Sample collection

Sample collection included (i) bulk soil samples to characterise the soil types within the SWAT model database; and (ii) surface samples to characterise possible sources and target sediments from the Barasona reservoir for the fingerprinting procedure.

Bulk soil samples used to derive SWAT model input data comprised 166 individual samples, two per sampling point, which were combined in the field to form 83 composite samples. They were collected with a core sampler at soil depths between 20-50 cm depending on the soil types.

Surface source samples to characterise potential sources for the fingerprinting procedure were collected across representative sites following a spatial distribution defined in open-source R software environment (Fig. 1a) after excluding areas, that in general, comprised massive rock outcrops with very little soil development and vertical slopes. Excluded areas comprised locations with altitude above 2000 m a.s.l. and slopes greater than 30%. The sampling definition followed a non-aligned random spatial sampling method implemented with the 'spsample' function from the sp library. This method generates a random spatial sample while preserving an even spatial distribution of points across the study area. Moreover, the number of the selected representative sites

for each source category was checked to ensure that they were balanced in relation to the percentage distribution of the main land uses/land covers in the Barasona catchment (Fig. 1c). Samples from the representative sites were taken in areas where there is high potential sediment yield connectivity from hillslope to channel and relatively easy access.

A total of 384 individual surface samples, 4 samples per sampling point, were collected by using a cylindrical core 5 cm long and 6 cm of diameter and combined in the field to form 96 composite samples (83 surface soil and 13 subsoil). The depth of sampling interval (5 cm) was selected because of the stoniness and high surface soil roughness in the study soils (Minella et al., 2004). 35 of the surface samples were from forest, 25 from agricultural fields and 23 from scrubland (Fig. 1a). Subsoil source samples comprised six composite samples from badlands on Eocene marls in the intermediate part of the catchment and another seven composite samples from other eroded areas. Forest sources included land uses within the forest land covers observed in the region, alpine grasslands and dense scrublands. The latter were included in the forest sources as they were thought to correspond with forest areas where burning practice to produce pastures for livestock had been common in the beginning of the past century. These land uses are referred to as 'forest' in the remainder of the paper. Scrubland sources included the rest of the scrublands and grasslands with low-intensity grazing. Agricultural sources are representative of the main cultivation practices in the catchment comprising annual production of rain-fed cereals (barley, wheat and sunflowers) with the combination of conservation and traditional tillage. Subsoil sources are representative of areas with clear evidence of active subsoil erosion (badlands included). Although channel banks are commonly sampled as a source for fingerprinting procedures, in this

study they were not sampled because erodible channel banks are either non-existent or poorly developed in some secondary streams.

Surface target sediments from the Barasona reservoir were collected in October of 2011, before the autumn rainfalls when the water level was low and most of the deposited sediments were exposed. Six locations along the longitudinal axis of the reservoir (Fig. 1a; samples A-F) and one more from its eastern branch (sample G) were selected to be representative of the most recent and accessible reservoir filling. For each location, four surface samples were collected at 2 cm depth to generate a composite sample. The sample depth could be considered as representative of around one hydrological year's deposit based on previous studies of reservoir filling (Valero-Garcés et al., 1999; Navas et al., 2004).

2.3 Sample analysis

All samples were initially oven-dried at 35 °C, gently disaggregated and sieved to <2 mm. For SWAT, the bulk samples were analysed to obtain- soil texture, bulk density and soil organic carbon. However, for the fingerprinting procedure, the surface samples were also sieved to <63 µm to isolate a comparable grain size fraction between source and sediment materials (e.g., Walling 2005; Smith and Blake 2014). Surface samples and target sediments were analysed to obtain the following tracer properties, or fingerprints: textural sizes (n=3); soil organic carbon (TOC); magnetic susceptibility (n=2); the elemental composition (n=25) and the mass activities of environmental radionuclides (n=6). For detailed explanation of the analytical methods of grain size, TOC and magnetic susceptibility see Palazón et al. (2015b)

The analysis of the total elemental composition was carried out after total acid digestion with HF (48%) in a microwave oven (Navas and Machín, 2002). Samples were analysed for the following 28 elements: Li, K, Na (alkaline), Be, Mg, Ca, Sr (light metals), Cr,

Cu, Mn, Fe, Al, Zn, Ni, Co, Cd, Tl, Bi, V, Ti and Pb (heavy metals), B, Sb, As (metalloids), and P, S, Mo and Se. Analyses were performed in triplicate by inductively coupled plasma atomic emission spectrometry with a Perkin Elmer OPTIMA 3200 DV ICP-AES and resulting concentrations expressed in milligrams per kilogram. Those elements returning measurements below the detection limit (Co, Cd and Se) were excluded.

Radionuclide activity concentrations in the samples were measured using a Canberra high-resolution, low background, hyperpure germanium coaxial gamma detector model XtRa GX3019. The detector had a relative efficiency of 50% and a resolution of 1.9 keV (shielded to reduce background) and was calibrated using standard samples that had the same geometry as the measured samples. Subsamples of 50 g were loaded into plastic containers (Navas et al., 2005a, b, 2014). Count times over 24 h provided an analytical precision of approximately ± 3 –10% at the 95% level of confidence. Gamma emissions of ^{238}U , ^{226}Ra , ^{232}Th , ^{40}K , ^{210}Pb and ^{137}Cs (expressed in Bq kg^{-1} air-dry soil) were measured in the samples (Table 2) with appropriate corrections for laboratory background (Palazón et al., 2015a). Moreover, the fallout radionuclide termed unsupported or excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) was estimated to distinguish it from the *in situ* supported ^{210}Pb component (Gaspar et al., 2013; Mabit et al., 2014).

Table 2.- Photopeaks used to measure radionuclide activities.

Radionuclide	Photopeak (keV)
^{238}U	63
^{226}Ra	352 (Van Cleef 1994)
^{232}Th	911
^{40}K	1461
^{210}Pb	47 (Appleby and Oldfield, 1992)
^{137}Cs	661.6

2.4 SWAT model assessment

The SWAT model is a continuous and physically based, semi-distributed, agro-hydrological model, which operates on a daily time step (as a minimum) and at the catchment scale. This is designed to predict the impact of management on water, sediment, and agricultural chemical yields in ungauged catchments (Arnold et al., 1998). Theory and details of the different processes integrated in the SWAT model (Neitsch et al., 2010) are available online in SWAT documentation (<http://swatmodel.tamu.edu/>).

SWAT discretises the catchment into Hydrological Response Units (HRUs) by distinctive combinations of categorized land uses, soil types, and slope. For this process the study area was characterised using: (i) a derived slope map based on a digital elevation model (Fig. 1b, DEM; 25x25m) obtained from the National Geographic Institute (IGN 2011); (ii) a land use map that was extracted, edited and resampled from the European Project Corine Land Cover map (CLC2000); and (iii) the Digital Soil Map of Aragón at a scale of 1:500,000 (Fig.1b, Soil Map of Aragón, Machín, unpublished data, 2000). The overlapping of these spatial layers resulted in 5,399 HRUs (see details in Palazón and Navas, 2014).

The land cover categories identified in the CLC2000 legend were reclassified to assign an equivalent class present in the SWAT2009 database. Soil properties were characterised in a user soil database which was developed with the information from the soil types and incorporated within the SWAT2009 soil database. The information required by SWAT for soil types includes the soil hydrologic group, soil texture, bulk density, available water capacity, saturated hydraulic conductivity, organic carbon content, soil depth, USLE soil erodibility (K) factor and albedo. The soil parameters were defined based on the samples collected in the catchment (totalling 215, including

data from previous studies) (Table 3). The information about soil texture, bulk density and organic carbon content was extracted from the analysis of the soil samples. The soil hydraulic properties, available water capacity and the saturated hydraulic conductivity were calculated with hierarchical pedotransfer functions from the ROSETTA model (version 2.1 2002, Schaap et al., 2001) based on the soil texture and bulk density. The USLE soil erodibility (K) factor was calculated according to a general equation developed by Williams (1995) that was recommended in Neitsch et al. (2011). Soil depths were defined based on field observations (Palazón and Navas, 2014). The albedo of the soil types was based on the descriptions of Tsvetsinskaya et al. (2002) for different soil types.

Climate inputs of daily minimum and maximum temperature and rainfall data in this SWAT project were based on measured historic data within or close to the region. Thus, rainfall records from four stations were selected from a revised database (1955–2006; Vicente-Serrano et al., 2009), and daily temperature records from five stations were obtained from the State Meteorological Agency (AEMET, Fig. 1a). To account for temperature and precipitation gradients according to the relief, ten homogeneous elevation bands and their lapse rates in height were defined in each subcatchment.

Table 3.- Soil properties included in SWAT database for the soil types present in the Barasona catchment following FAO (2007) classifications.

Predominant soil (secondary soil)	Organic carbon	Clay	Silt	Sand	Rock fragment	Bulk density	Soil erodibility (K) factor	Available water capacity	Saturated hydraulic conductivity	Soil albedo
	%	%	%	%	%	g cm ⁻³	*	mm H ₂ O/mm soil	mm/h	-
Cambisol (Kastanozem)	2.7	8	48	44	38	1.34	0.14	0.17	17.62	0.07
Calcaric Cambisol	1.3	19	78	3	47	1.39	0.36	0.22	6.74	0.04
Dystric Cambisol (Umbric Leptosol)	2.5	25	72	3	52	1.3	0.3	0.23	7.43	0.04
Dystric Cambisol (Umbric Lithic Leptosol)	2.4	16	80	4	82	1.2	0.32	0.25	17.88	0.08
Fluvisol	2	15	74	11	60	1.2	0.25	0.25	23.67	0.12
Kastanozem	1.1	12	48	40	20	1.4	0.17	0.17	10.19	0.12
Kastanozem (Rendzina)	2.1	13	38	49	44	1.12	0.14	0.21	33.91	0.09
Kastanozem (Rendzina + Haplic Kastanozem)	2.2	21	73	6	32	1.2	0.28	0.25	15.41	0.1
Kastanozem (Rendzina + Lithic Kastanozem)	4.3	22	74	4	45	1.1	0.3	0.26	20.68	0.2
Lithic Kastanozem	2.7	12	48	40	12	1.4	0.45	0.17	10.19	0.12
Leptosol	3.1	21	69	10	65	1.4	0.23	0.22	6.76	0.2
Rendzina	1.7	9	32	59	72	1.18	0.14	0.18	45.6	0.2
Lithic Rendzina	2.4	20	59	21	30	1.4	0.16	0.21	6.65	0.04
Chromic Luvisol	2.6	13	57	30	58	1.14	0.15	0.25	34.26	0.04
Calcaric Phaeozem	1.9	20	22	58	29	1.38	0.13	0.16	13.78	0.12
Haplic Phaeozem	1.9	11	59	30	24	1.13	0.16	0.25	40.87	0.12
Haplic Phaeozem (Leptosol)	2.1	12	59	29	34	1.19	0.15	0.24	29.3	0.12
Regosol	1.9	20	60	20	54	1.26	0.17	0.23	13.74	0.2

*units: 0.031 (metric ton m² hr)/(m³-metric ton cm)

The SWAT model was calibrated (period 2003-2006) and validated (period 1994-2002) for hydrology and sediment yield in Palazón and Navas (2014). For monthly streamflow simulations, the periods yielded Nash–Sutcliffe efficiency coefficients (Nash and Sutcliffe, 1970) greater than 0.7 and 0.5, for calibration and validation, respectively. Simulated sediment supply to the reservoir for both periods compared well with the specific sediment yields obtained by bathymetric survey (Avendaño-Salas et al., 1997) and other estimates of sediment production in the region (e.g.: Alatorre et al., 2010; López-Tarazón et al., 2012). Furthermore, simulated sediment production from the individual land uses and covers were in agreement with values from the literature (Palazón and Navas, 2016). Therefore, the outputs of this study were used here to assess the spatial distribution of the annual sediment production for the period 2003-2005. This period was selected as it compiles three years with different annual precipitation (Table 4). Within the study period, 2003 was a wet year (1383 mm) while the other years were drier (871-882 mm). The SWAT model was not calibrated for erodible riverbanks because they are rocky, stony or composed of block deposits. Therefore the model default option was selected and erosion in the riverbanks was assumed to be negligible. The sediment production was also assessed by grouping the land uses and land covers in four main sources: agricultural land, forests, scrublands and badlands. Moreover, the percentage of the relative sediment production from each source was determined relative to the respective surface area in the catchment in order to compare with the results from the fingerprinting approach.

Table 4.- Climatic and simulated surface runoff data for the period 2003-2005

		mean	sd	2003	2004	2005
(°C)	Temperature	9.8	0.4	10.2	9.8	9.4
(mm)	Precipitation	1045	292	1383	882	871
	Surface runoff	114	50	172	86	85

2.5 Fingerprinting procedure

After exclusion of non-conservative fingerprint properties a range test between sources and sediment tracer properties was applied (Martínez-Carreras et al., 2010a; Smith and Blake 2014; Palazón et al. 2015a). The ability of the remaining properties to discriminate sources was investigated by conducting the commonly used combination of statistical tests proposed by Collins et al. (1997). The nonparametric Kruskal–Wallis H test was first used to identify fingerprint properties which exhibited significant differences between sources followed by a stepwise discriminant function analysis (DFA) based on the minimization of Wilks' lambda which determine the minimum optimal group of tracers. The lambda value approaches zero as the variability within sources is reduced relative to the variability between source groups. The resulting optimum composite fingerprint comprises the minimum number of tracers that provide the greatest discrimination between the sources.

The relative apportionment of each potential source was assessed by a mixing model which solved the system of linear equations defined by a conservative mass balance. Within this, the sources, characterised by the optimum composite fingerprint, were multiplied by its unknown source apportionments and summed to be equal to the same equivalent tracers from the mixture sediment samples. The system has to satisfy two constraints: that the apportionments must lie between 0 - 100 % and sum to 100 %. In this study the predicted relative apportionments from each mass balance equation were solved by an optimization procedure which minimises an objective function, or goodness of fit (GOF, based on Collins et al. 1997), defined by:

$$GOF = 1 - \frac{1}{n} \times \sum_{i=1}^n \left\{ \left(\frac{\left| b_i - \sum_{j=1}^m x_j a_{i,j} \right|}{\Delta_i} \right)^2 \right\}$$

where b_i is the value of tracer property i ($i=1$ to n) in the sediment sample, $a_{i,j}$ is the mean concentration of tracer property i in source type j ($j=1$ to m), x_j is the unknown relative weighting contribution of source type j to the sediment sample, m is the number of potential source types, n is the number of tracers of the optimum composite fingerprint, and Δ_i is used as correction factor to normalize the tracer properties ranges.

A Monte Carlo global sampling routine designed to provide the optimal solution after a uniform exploration of the entire parameter space was adopted (Palazón et al., 2015b) and the value of each fingerprint was randomly modified with an iterative procedure according to its Student's t distribution defined by the mean and standard deviation values of each source group. A large number of possible solutions (10^6) were generated, tested and ranged by their GOF value to extract the 100 solutions that were found to best fit the objective function (GOF closer to 100%). Therefore, the optimal solution was characterised by the mean and the standard deviation of the extracted source apportionments and the lowest GOF value achieved by the optimization procedure for the selected solutions. Moreover, the 100 solutions used to characterize the optimal solution enabled examination of frequency distributions of the extracted apportionments to assess the distributions of the solutions. Furthermore, following Palazón et al. (2015b), an auto-evaluation method using 1000 virtual mixture samples generated by their Student's t distribution was applied to test the accuracy of the multivariate mixing model by the root mean squared error (RMSE).

3. Results

3.1 Sediment production by SWAT model

For the assessed years 2003-2005, the simulated average specific sediment yields by HRUs (SSY: $\text{t ha}^{-1} \text{ year}^{-1}$) showed great variability across the catchment (Fig 2). The SSY had a mean value of $0.96 \text{ t ha}^{-1} \text{ year}^{-1}$ and a standard deviation of $9.54 \text{ t ha}^{-1} \text{ year}^{-1}$. Despite this variability, more than 50 % of the HRUs produced SSY lower than $0.01 \text{ t ha}^{-1} \text{ year}^{-1}$ and only 3 % of the HRUs generated SSYs exceeding $2 \text{ t ha}^{-1} \text{ year}^{-1}$. The highest SSYs were concentrated in the intermediate part of the catchment containing badlands. Apart from these areas of greatest sediment production, i.e. relatively high SSY with values between 1 and $2 \text{ t ha}^{-1} \text{ year}^{-1}$, were located mostly in the lowlands, coinciding with agricultural land use, and also dispersed in the uppermost part of the catchment. The distribution of the averaged SSY showed greater SSY for the northern part of the catchment than for the lowlands.

The simulated SSY for the assessed groups of agricultural land, forests, scrublands and badlands, yielded different results for the groups and for the years (Fig 3). Apart from the badlands, the agricultural land yielded the highest SSY with mean values of SSY almost one order of magnitude greater than the scrubland and forest land uses (Table 5). Scrubland and forest yielded similar low SSY values which were the lowest for forest. The badlands yielded the highest SSY values, which, at a minimum, were one order of magnitude greater than the agricultural land, and unlike the other land uses they had minimum values considerably greater than 0. Apart from forest, the other land uses and land covers yielded highest SSY values for the year 2003, lower for the year 2004 and the lowest SSY was for the year 2005. All land uses and land covers yields almost doubled SSY in year 2003 in comparison to year 2005. In general, as for the whole catchment, higher SSY values were observed for those which are further north than for the southern ones.

Fig. 2 Averaged specific sediment yield (SSY: t ha⁻¹ year⁻¹) for the period 2003-2005 simulated for the Barasona catchment with the zoom of the main badlands area.

Fig. 3 Annual specific sediment yield (SSY: $\text{t ha}^{-1} \text{ year}^{-1}$) for the four assessed sources for the years 2003, 2004 and 2005 simulated for the Barasona catchment.

Table 5.- Annual specific sediment yield ($\text{t ha}^{-1} \text{ year}^{-1}$) by agricultural, forest, scrubland and badlands groups for the HRUs simulated by SWAT for the study period (2003-2005).

Agricultural	n=726				
	mean	median	sd	max	Min
2003	1.16	0.23	2.12	17.60	0.00
2004	0.83	0.07	1.80	17.87	0.00
2005	0.57	0.06	1.10	8.32	0.00
Forest	n=2821				
	mean	median	sd	max	min
2003	0.06	0.00	0.30	7.44	0.00
2004	0.01	0.00	0.04	0.77	0.00
2005	0.01	0.00	0.06	1.47	0.00
Scrubland	n=1471				
	mean	median	sd	max	min
2003	0.18	0.03	0.47	6.20	0.00
2004	0.04	0.00	0.11	1.45	0.00
2005	0.04	0.00	0.11	1.10	0.00
Badlands	n=56				
	mean	median	sd	max	min
2003	135.84	118.54	96.09	440.44	15.81
2004	53.14	42.65	40.10	191.58	5.38
2005	41.94	36.11	27.87	117.35	4.81

3.2 Fingerprinting sediment contributions to the reservoir sediment

The fingerprinting approach for the Barasona catchment was based on analysis of contributions from four possible sediment sources: agricultural, forest, subsoil and scrubland. Following the recommendations of Granger et al. (2007) and Koiter et al. (2013), TOC, P and grain size fractions were excluded from the analyses as they were considered non-conservative properties. The range test resulted in the exclusion of the magnetic frequency dependence property (χ_{FD}) for the following analysis because its minimum value for the reservoir sediments is lower than the minimum value in the source samples. The Kruskal-Wallis H-test resulted in the identification of ^{40}K , ^{137}Cs ,

$^{210}\text{Pb}_{\text{ex}}$, ^{232}Th , χ_{LF} , As, B, Ca, Cu, K, Li, Mg, Ni, Pb, Se, S, Sr, Ti and Zn which showed significant differences between the four sources at the 5 % confidence level. From the 19 remaining tracer properties passing the previous steps, DFA led to the selection of ^{40}K , ^{137}Cs , Li, Sr and Ti (Table 6 and 7), as the optimum composite fingerprint. The DFA achieved a Wilks' lambda value of 0.18 and a source discrimination of 86.5% of sources correctly classified. The first two discriminant functions calculated by the DFA from the stepwise properties for the selected four sources are depicted in Figure 4. Source sample clusters overlapped between agricultural, scrubland and forest sources and also between scrubland and forest sources.

Fig. 4 Two-dimensional scatter plot of the first and second canonical discriminant functions from the stepwise discriminant function analysis (DFA).

Clear differences were observed between the properties that were selected as optimum composite fingerprint for the different sources (Table 6 and Fig. 5). Subsoil showed the greatest differences in measured tracer property concentrations selected as part of the optimum fingerprint. The tracers exhibiting the largest differences between sources were Sr and ^{137}Cs .

Table 6.- Source means and reservoir values of the tracer properties selected as optimum composite fingerprinting for the four potential sediment sources and the Barasona reservoir sediment samples, respectively.

		^{137}Cs Bq kg ⁻¹	Sr mg kg ⁻¹	^{40}K Bq kg ⁻¹	Ti mg kg ⁻¹	Li mg kg ⁻¹
Sources	Agricultural (n=25)	13.0	145.6	587.2	3654.8	46.9
	Forest (n=35)	81.7	107.5	567.9	4347.4	47.7
	Scrubland (n=23)	33.9	201.4	513.6	4011.3	46.1
	Subsoil (n=13)	0.9	460.7	657.5	2964.6	60.1
Reservoir sediments	Sample A	5.9	270.2	892.7	3190.0	51.5
	Sample B	0.0	321.0	760.0	3280.0	55.3
	Sample C	0.0	321.3	787.0	3230.0	45.6
	Sample D	0.8	298.6	562.0	3060.0	40.7
	Sample E	0.4	326.4	744.0	3120.0	48.7
	Sample F	0.0	238.6	647.0	3120.0	43.5
	Sample G	1.8	145.0	528.0	3140.0	34.3

Table 7.-Results of the stepwise discriminant function analysis to identify the optimum composite fingerprint.

Fingerprint property added	Wilks' lambda
^{137}Cs	0.452
Sr	0.294
^{40}K	0.234
Ti	0.206
Li	0.180

Fig. 5 Box plots of the tracers selected as the optimum composite fingerprint for the agricultural, forest, scrubland and subsoil sources.

The mixing model incorporated all tracer properties that were selected by the DFA as the optimum composite fingerprint to solve the mass balance equation for the sediment mixture samples and the virtual mixture samples. The auto-evaluation of the procedure with virtual mixture samples provided a RMSE of 9 %. The standard deviation of the best combination allowed us to compare and assess the solution dispersion as large values indicate poor source contribution ascription. The outputs appeared to be stable from different random number seeds supporting the performance of the optimization

procedure and the convergence of the analysis. In each repetition, the solution dispersion was less than 20 % of its mean value (Table 8). Forest and scrubland produced the lowest standard deviations, whereas, agricultural and subsoil sources yielded higher standard deviations which were highest for the mixture sample F. For all target samples, the frequency distributions of the optimal solutions showed narrow, symmetrical and unimodal distributions for forest sources and in most cases for scrubland sources (Fig. 6). However, the frequency distributions of the solutions for agricultural and subsoil sources showed more dispersion with skewed patterns and in some cases bimodal and multimodal distributions, as for samples D and F.

Table 8.- Mean percentages of GOF, source contributions and their standard deviations (in parentheses) obtained as optimal solutions from the mixing model for forest, agricultural, scrubland and subsoil sources to the Barasona sediment mixture samples.

	GOF	Forest	Agricultural	Scrubland	Subsoil
Sample A	98.9	5.2 (3.8)	15.7 (12.1)	6.4 (5.4)	72.7 (11.2)
Sample B	99.7	2.3 (1.9)	14.7 (9.7)	5.2 (4.0)	77.9 (9.0)
Sample C	99.5	3.0 (2.3)	16.1 (11.7)	5.2 (4.9)	75.7 (11.0)
Sample D	99.6	2.0 (1.9)	30.3 (15.8)	6.9 (6.0)	60.8 (13.0)
Sample E	99.7	2.4 (2.2)	14.1 (9.4)	5.3 (4.2)	78.3 (7.9)
Sample F	99.7	1.6 (1.5)	40.1 (19.7)	4.4 (4.0)	53.8 (18.2)
Sample G	99.2	1.9 (1.8)	84.0 (12.9)	5.9 (4.9)	8.2 (11.0)

Mean contributions from agricultural, forest, scrubland and subsoil sources varied between sediment samples in the reservoir (Fig 6 and Table 8). The source proportions were low (< 7%) and similar for both forest and scrubland sources and greater for agricultural land and subsoil, with a general predominance of subsoil sources in the reservoir mixtures. The largest differences in source contributions were obtained for the sample G with apportionments greater than 80 % from the agricultural sources instead of subsoil sources. Sample F also differed from the others with similar contributions from agricultural and subsoil sources.

Fig. 6 Distribution along the Barasona reservoir of the frequency distributions of the 100 solutions which comprised the optimal fingerprinting solutions in pie charts obtained with the mixing model for the mixture samples.

3.3 Results of the combined procedures: SWAT and fingerprinting

The results from SWAT model and sediment fingerprinting had different characteristics. The SWAT model simulated spatial and timescale specific sediment yields from the main four land use/land cover groups whereas the sediment fingerprinting procedure

generated relative source apportionment values for surface sediments accumulated in the reservoir. Therefore, to compare the two procedures, the sediment production from each land use/cover was assessed relative to the area of each land use/cover based on mean simulation results for the 3 year study period (2003-2005). Thus, the relative sediment production obtained from SWAT outputs could be directly compared with the relative source apportionments obtained from the fingerprinting procedure for the reservoir samples. Standardised data from the combined procedures were in agreement with similar outcomes for the 6 reservoir samples A-F (Table 9).

Table 9.- Comparison between simulated sediment productions by SWAT and range of fingerprinting source apportionments obtained for the reservoir samples (A-F).

Source group	Modelled sediment production	Range of apportionment
Agricultural	14.5	14.1 - 40.1
Forest	2.1	1.6 - 5.2
Scrubland	2.6	4.4 - 6.9
Badlands and subsoil	80.8	53.8 - 78.3

4. Discussion

The SWAT model results showed the variable distribution of SSY in the catchment related to the combined effect of the land use, slope gradient and soils. The limited percentage of HRUs that produced more than $2 \text{ t ha}^{-1} \text{ year}^{-1}$ was closely linked with the badlands, thereby supporting the importance of this source of sediment in the catchment, as was also reported by other authors using different methods (e.g. Martinez-Casasnovas and Poch, 1998; Alatorre et al., 2010; Brosinsky et al., 2014; López-Tarazón et al., 2015). The badland HRUs achieved high to extreme sediment production

levels, which were outliers in the distribution of simulated values. As a secondary source of sediment in the catchment, high levels of simulated sediment were found in agricultural lands. Differences in sediment production between the assessed land use/land covers were mostly related to their vegetation covers which affected erodibility. As no vegetation cover protects the badlands, sediment production here was related only to the availability of material to be eroded by the precipitation. The greater SSY in 2003 compared to the other years reflects the higher precipitation in this period. Moreover, greater SSY for the northern part of the catchment was related to the observed precipitation and topographic gradients in the catchment (Palazón and Navas, 2016).

Within the fingerprinting procedure, source sample properties were found to overlap to some extent based on the results of the DFA, which explains why the analysis did not achieve 100 % correct classification of source groups. This was most notable between agricultural and scrubland sources, and also between the latter and forest. This could represent the transitional states of the natural reforestation that occurred in the catchment after abandonment of agricultural lands and the progressive conversion of agricultural areas into natural forest (Navas et al., 2008; Lasanta and Vicente-Serrano, 2012). This is meaningful in the context of the known catchment history of land abandonment since the 1950s.

Tracer discrimination between sediment sources demonstrated the importance of the fallout radionuclide ^{137}Cs as a discriminator wherein it exhibited the largest differences between sediment sources. Large differences in Sr content in subsoil sources, which almost doubled those in the other land uses, were likely related to the nature of the mineral composition of the substrate due to the contribution of parent materials (Navas and Machín, 2002). Lower Ti content in subsoil samples in comparison with the other

sources might reflect the effects of weathering. As Ti minerals are very stable, the loss of some clay-size layered silicates might relatively enrich Ti content in the upper soil horizons (Kabata-Pendias and Pendias, 2001). Therefore, well developed and undisturbed soils could have their topsoil more enriched in Ti than eroded ones. Lower differences between sources were found for Li and ^{40}K with higher amounts in the subsoil source that could be related with the common contents of both elements in sedimentary rocks with argillaceous materials (Kabata-Pendias and Pendias, 2001) which are the dominant lithology of the substrate in the subsoil sources.

The fingerprinting apportionment results were consistent with the conceptual understanding of the catchment and also with the outputs from SWAT model. The RMSE obtained by the auto-evaluation was moderate suggesting that while the sediment fingerprinting procedure can discriminate between the selected sources, natural revegetation represents an intermediate stage in the natural plant succession which can reduce clarity in overall source fingerprint differences. Hence sources could be confused and interchanged within the best solution for the mixing model leading to increased uncertainty. The high dispersion showed by the frequency distributions of some sources could also be related with the moderate accuracy of the procedure. For example, the bimodal distribution for agricultural and subsoil sources showed that variations of their contributions could also correspond to possible optimal solutions. Furthermore, despite the RMSE obtained for the fingerprinting approach, findings by Palazón et al. (2015b) in relation to different fingerprinting procedures yielding different source apportionment outcomes, suggest that further research is needed to explore mixing model performance for different optimum composite fingerprints provided by different selection procedures.

Based on the RMSE obtained by the auto-evaluation test, confidence in around 91 % of source ascriptions was underpinned for this case study. The results of the auto-evaluation were in accord with the observed agreement between the source contributions obtained with the mixing model for the reservoir samples and results from previous studies in the region (e.g. Alatorre et al., 2010; Palazón et al., 2015a; Palazón and Navas, 2016) for which the main and secondary sediment sources are badlands and agricultural lands, respectively. Variations in the source contributions from the tail to the mouth of the reservoir could be related to the surroundings of the reservoir. For example, this may explain a decrease in the relative subsoil contribution to reservoir sediments between the Ésera River mouth and the reservoir tail. This effect is especially important for the part of the reservoir that collects the discharge of the Sarrón stream (sample G) which drains a small catchment area mostly covered with agricultural land in which badlands are not present.

Although the characteristics of the results from the SWAT model and sediment fingerprinting were different they offer complementary insights on the erosion processes in the Barasona catchment. The model provided information related to the distribution and timescale quantification of source sediment production based on the physiographic characteristics of the discretised HRUs (land use, soil type and slope range) influenced by the continuous simulated climate for the study period. Total sediment yield to the reservoir could be obtained from the model outputs but not the source contributions to this sediment yield. Whereas the fingerprinting procedure provides information on the relative source contributions to the surface sediments accumulated in the reservoir. Both procedures were consistent with the conceptual understanding of the catchment which pointed to the badlands or subsoil and agricultural sources as the main and secondary sediment sources, respectively. The differences between the averaged

simulated SSY for the selected representative period (2003-2005) for the assessed land uses and land covers compare well to the source apportionments obtained for the reservoir mixture samples, which are estimated to represent around one to two years based on sedimentation rates in the reservoir calculated by Valero-Garcés et al., (1999) and Navas et al., (2004). Therefore, the results of the combined procedures enabled us to derive information about the transport capacity of the river system from the sources to the reservoir. The relative source apportionments compared well with the simulated sediment productions suggesting that the river system has sufficient capacity to transport the eroded sediments.

The slightly greater contributions in the reservoir sediments from scrublands in relation to forests were comparable to similar simulated sediment productions. However, the greater extent of forests, suggest that sediment produced from scrublands could be more efficiently transported by the river system than sediment from forests. This could also be related to higher connectivity as well as to the distribution of scrublands in the catchment, as they mostly occupy locations closer to streams than forests. Similarly, contributions from agricultural and subsoil/badlands sources suggest that the spatial distribution and proximity to main streams and the reservoir of the agricultural source was an important factor in its greater proportional contribution to reservoir sediments compared to its simulated sediment production. The largest sediment contribution of the badlands occurred despite their limited spatial extent in the catchment and was related to the high sediment availability and production rates together with the location of eroded material in alluvial fans that are closely connected to the streams and, therefore, efficiently transported through the hydrological network over the temporal scale of the study.

A number of potential limitations should be taken into account when interpreting the findings of this study. Sampling in the reservoir was only undertaken for a single campaign due to available funding and catchment scale and access limiting the observation to a single year of sediment deposition. Even so, the fingerprinting results were in accordance with previous studies in the catchment, more sampling campaigns (yearly or seasonally) could show temporal changes in catchment source contributions which could also be related with modelling outputs to derive information related to seasonal sediment transport processes or different climate conditions of the years. Although fingerprinting results for the reservoir could be considered representative for this large catchment, additional sediment samples along the rivers assessed with the fingerprinting procedure could enhance knowledge of source contributions delivered through the system to the Barasona reservoir. Further research is needed to understand the role of varying sediment source contributions and upstream erosion on sediment dynamics within the catchment. Despite the above limitations, previous work at the small headwater catchment scale (Palazón et al., 2014) and this study at the large catchment scale demonstrate that the combined procedures provide useful information related to sediment connectivity of catchment systems from sources to downstream sediment mixtures. Such a combined approach is recommended for applications in other environments on the basis of its ability to provide insights into the functioning of sediment generation and transport within catchments.

5. Conclusions

The application of the SWAT model to the alpine-prealpine Barasona catchment provided important insights into the spatial distribution of sediment yields from agricultural, forest, scrubland and badlands sources under different annual precipitation

conditions. Results showed that sediment production was related to the interplay between land use, slope, precipitation amount and gradient. Simulated sediment production between source categories was highest for badlands, intermediate for agricultural lands and low for forest and scrubland.

Frequency distributions of optimal sediment source fingerprinting solutions showed, in most cases, similar patterns. Barasona reservoir bed surface sediment mixtures gave consistent results in this regard apart from one area where localised deposition was related to discharge from a small catchment where badlands were absent. Based on the combined procedures, the river network appeared to efficiently transport sediment eroded in the catchment over the timescale of the study. Subsoil sources were, together with agricultural sources, the most important contributors to reservoir siltation in the catchment, with higher contributions from subsoil sources in most of the cases, whereas forest and scrubland sources did not contribute significantly.

Results obtained from the SWAT model and the fingerprinting procedure provided two different but compatible approaches to study the processes involved in reservoir siltation and associated threats to water and energy security. The combination of procedures produced new information on sediment production from land uses and the yields to the reservoir as well as improving knowledge of erosion processes and sediment delivery to support management actions. Furthermore, the agreement obtained between the approaches used in this study increases the confidence that can be attributed to outputs from both the catchment modelling and fingerprinting procedures to support sediment management at the catchment scale. The success of the combined modeling and fingerprinting procedure to gain information related to erosion processes in this mountain catchment (with a scarcity of data, large climatic gradients and an impounded river), affords an opportunity to extend the use of this combination of

procedures to further catchments in the Pyrenean region and other mountain environments.

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