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1 FingerPro: An R package for tracking the provenance of sediment
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9 ABSTRACT

10 Soil loss by erosion processes is one of the largest challenges for food production and 11 reservoir siltation around the world. Information on sediment, nutrients and pollutants is 12 required for designing effective control strategies. The estimation of sediment sources is 13 difficult to get using conventional techniques, but sediment fingerprinting is a potentially 14 valuable tool. This procedure intends to develop methods that enable to identify the 15 apportionment of sediment sources from sediment mixtures.

We developed a new tool to quantify the provenance of sediments in an agroforest 16 catchment. For the first time, the procedure for the selection of the best combination of 17 18 tracers was included in the tool package. An unmixing model algorithm is applied to the 19 sediment samples to estimate the contribution of each possible source. The operations are compiled in an R package named FingerPro, which unmixes sediment samples after 20 selecting the optimum set of tracers. An example from a well-studied Mediterranean 21 22 catchment is included in the package to test the model. The sediment source apportionments are compared with previous results of soil redistributions where ¹³⁷Cs 23 derived rates validate the unmixing results, highlighting the potential of sediment 24 fingerprinting for quantifying the main sediment provenance. Fingerprinting techniques 25

26 will allow us to better comprehend sediment transport to water ecosystems and reservoirs

and its detrimental effect on the quality of the water and aquatic habitats.

The **FingerPro** package provides further understanding of the unmixing procedure through the use of graphical and statistical tools, offering a broader and easier application of the technique.

31 KEYWORDS: FingerPro, unmixing model, sediment source fingerprinting, source
 32 variability, R package

33

34 1. INTRODUCTION

35 Reliable information on sediment loads transported by a river or stream is crucial to evaluate the severity of reservoir siltation and river pollution. However, determining 36 sediment provenance or sediment budgets in catchments using conventional monitoring 37 38 techniques is often challenging. However, in most situations, it can be provided by applying tracing techniques. Fingerprinting techniques can be used to recognise sediment 39 40 sources and to determine their relative contribution, thereby allowing the identification of areas or land uses prone to erosion processes (Schuller et al. 2013). Soil erosion and 41 42 subsequent sediment transport are related to the loss of nutrients and their distribution in 43 the catchment (Lizaga et al. 2019). To assess this issue, several software and indices have been developed to quantify the effects of different erosion mechanisms, such as 44 connectivity (Shore et al. 2013), runoff (dos Santos et al. 2017), sheet and rill (Molnár 45 and Julien, 1998), wind erosion (Schmidt et al., 2017) and the subsequent effects on water 46 47 quality (Panagopoulos et al. 2011; Foucher et al. 2020). However, sediment source 48 fingerprinting has been developed in recent decades for catchment sediment and pollutant 49 investigation as the most powerful tool to assess this problem. The procedure identifies

sediment provenance and estimates the relative contribution of each potential source,using the selected tracer properties.

The first fingerprinting approach dates back to the seventies, based on mineralogical and grain size characterisation (Klages and Hsieh, 1975). The earliest fingerprinting studies were fundamentally qualitative in their result, but the introduction of quantitative mixing models was a methodological advance that enabled researchers to obtain quantitative results of the relative contribution from different sediment sources (Collins et al. 1997). Since these early works, sediment source fingerprinting applications have been greatly expanding with the development of new techniques (Owens et al. 2016).

59 The traditional approach for applying source-tracing methods is to define the relevant 60 tracer properties that provide a particular signature between all source samples and 61 unequivocally discriminate the different sources (Collins and Walling, 2002). Due to the 62 inherent complexity of catchment characteristics, with large variations in climate, geology, land use, vegetation, soil, and management practices, commonly, no unique 63 64 tracer can discriminate between multiple sediment sources. Consequently, different tracer properties need to be analysed, such as radionuclides (Wallbrink et al. 1998; Evrard et al., 65 66 2020; Navas et al., 2020), geochemistry (Martínez-Carreras et al. 2010; Smith and Blake, 67 2014; Gaspar et al., 2019a), ultraviolet-visible spectra derived tracers (Ramon et al., 2020), CSSI (Reiffarth et al. 2019) and eDNA (Evrard et al. 2019). 68

The fundamental theory that supports this technique is that the tracer properties of the sediment mixtures are directly comparable to the sediment of the sources. A common procedure, the so-called "range test", checks if sediment tracers are conservative excluding the tracers of the mixture/s outside the minimum and maximum values in the potential sediment sources. This procedure prevents the inclusion in the optimum tracers of the fingerprint properties exhibiting non-conservative behaviour. However, the 75 exclusion of a great number of fingerprint properties likely suggests that not all sources 76 have been correctly identified or characterised. Thus, the methodology for tracer selection 77 is an open question that is being discussed at present by several authors since different 78 tracer selection methods could lead models to different results (Pulley et al. 2015; Lizaga et al. 2020). Following this assumption, the two-stage statistical procedure previously 79 80 proposed by Collins and Walling, (2002), is commonly used to assess this 81 conservativeness. Thus, the Kruskal Wallis H test (KW) and discriminant function analysis (DFA) test the ability of individual tracers to discern between sediment sources 82 and select the best combination of tracers. This procedure was used to select the smallest 83 84 combination of tracers that provided the maximum discrimination of the identified source 85 categories and it is implemented by several authors as a common procedure when using 86 frequentist (Palazón et al. 2015; Lin et al. 2015; Gholami et al., 2020) and Bayesian 87 (Koiter et al. 2013; Barthod et al. 2015) unmixing models. Subsequently, the relative contribution of each identified source is estimated using a linear multivariate unmixing 88 89 model.

90 Due to the growing use of fingerprinting methods, other unmixing models, such as SIFT (Pulley and Collins, 2018), MixSIR (Moore and Semmens, 2008) and IsoSource 91 92 (Phillips and Gregg, 2003), appeared in the last years for pollution and ecological purposes. However, due to operational complexity and the need to use different statistical 93 software not included in the packages, the use of unmixing models is generally restricted 94 95 to academics with a good knowledge of the procedure. Refinement of the sediment source 96 fingerprinting techniques requires open-source models that help the user in tracer selection decisions and optimise this time-consuming process for non-expert and 97 academics with low programming and statistical skills by including the essential 98 99 statistical functions and plots.

To fill this gap, our objective was to develop for the first time an R package comprising an unmixing model with the additional tools needed to comprehend the effect of the selected properties on the model outcome. To this purpose, we create a new R package that combines in a novel assemblage the tools needed to unmix sediment samples and the previous statistical tests to select optimum tracers. Thus, we aim to provide an easy and straightforward way to apply the sediment fingerprinting technique aimed at beginners or non R users.

107 This paper presents the **FingerPro** package, a user-friendly application and freely 108 available software for users with limited or nor expertise in statistics. Thus, any user could 109 implement the fingerprinting procedure with limited previous experience in the technique 110 and with no need for additional software for statistical analyses. Furthermore, unlike 111 previous models, this new tool to identify sediment provenance has been successfully 112 tested with artificial samples (Gaspar et al. 2019b).

113 The analyses explained in this research are based on 1) a reproducible data set example 114 of small catchment included in the package and 2) another example of ongoing research 115 in a medium-size catchment to further describe the capability of the package. The medium 116 size catchment is selected as representative of mountain headwaters (South Pyrenean 117 region) that supply water to reservoirs as siltation and pollution is one of the main 118 environmental issues worldwide (Valero-Garcés et al. 1999). Through these two 119 examples, the utility of the **FingerPro** package for applying tools aimed at pre-processing input data or combining sources without significant differences before or after running 120 121 the unmixing model is shown. Through the provided examples, it is evident that fingerprinting methods are necessary to identify sediment sources to establish 122 123 management strategies for ensuring water supply to the lowlands while preserving water 124 quality.

125 **2. METHODS**

Sediment fingerprinting requires a preliminary analysis to select a subset of conservative tracers that discriminate the potential sources. Then, the relative contribution of each source is estimated using a linear multivariate unmixing model. This procedure is iterated considering the variability of the sediment sources to obtain the statistical distribution of the source contribution.

131 **2.1** Statistical analysis for the selection of tracer properties

132 Several statistical tests can be used to confirm source discrimination and select the optimal subset of conservative tracer properties, such as the procedure suggested by 133 134 Collins and Walling (2002). However, the use of many tests could remove a considerable number of tracers and therefore restrict the discrimination between sediment sources. 135 136 Consequently, none of the functions included in the **FingerPro** package are mandatory 137 and the tracer exclusions can be based on 'expert judgement' after visualising boxplots and results from the statistical tests included. The tracer selection methods implemented 138 139 in the package are:

i) Range test: the minimum and maximum values of the tracer properties in the
sediment sources are compared to those of the mixtures. The tracers falling out of the
range of the selected sources are removed from subsequent analyses. These properties
may not be conservative or their exclusion supports the existence of an additional hidden
source.

ii) Kruskal-Wallis H test: this is a rank-based nonparametric test used to determine if
there are significant differences between the medians of selected groups or sources. This
procedure removes tracers that do not show significant differences between at least two
of the sediment sources.

149 iii) Discriminant Function Analysis identifies the optimum set of tracers that 150 maximises the discrimination between the sediment sources whilst minimising the 151 number of tracers. This function executes a stepwise forward variable selection for 152 classification using the Wilk's Lambda criterion. The function selects the tracers based on 153 how much they decrease Wilks' lambda. At each step, the function includes the variable 154 that minimises the overall Wilks' lambda.

155 **2.2 Mixing model**

The relative contribution of each potential sediment source is determined using astandard linear multivariate mixing model:

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$$\sum_{j=1}^{m} a_{i,j} \cdot \omega_j = b_i$$

158 which satisfies:

160
$$\sum_{j=1}^{m} \omega_j = 1$$

161 $0 \le \omega_j \le 1$

where b_i is the tracer property i (i = 1 to n) of the sediment mixture, $a_{i,j}$ represents the tracer property i in the source type j (j = 1 to m), ω_j is the unknown relative contribution of the source type j, m represents the number of potential sediment sources and n is the number of tracer properties selected.

This system of equations is mathematically determined if the number of tracers is greater than or equal to the number of potential sources minus one $(n \ge m - 1)$. The procedure tries to find the source proportions that conserve the mass balance for all tracers from the complete exploration of the parameter space (Palazón et al. 2015). All possible combinations of each source contribution (0-100%) are examined in small increments, using Latin hypercube sampling (LHS) (McKay et al. 1979). The quality of each 172 candidate is measured using the following function or goodness of fit (GOF), based on173 the sum of squares of the relative error:

174
$$GOF = 1 - \frac{1}{n} \times \left(\sum_{i=1}^{n} \frac{|b_i - \sum_{j=1}^{m} \omega_j a_{i,j}|}{\Delta_i} \right)$$

where Δ_i is the range of the tracer property *i*, used as a normalisation factor. The combinations that reproduce the observed sediment mixture with the maximum GOF are selected as the solution.

178 **2.3 Variability analysis of the sources**

In small to large size catchments, the heterogeneity of sediment tracers, defined by different land uses, geomorphic processes, soil types or human activity, is always present. For this reason, fingerprinting studies should correctly characterise source variability by collecting several samples from each source. Thus, evaluation of the variability in tracer data used to characterise sediment sources is important to correctly interpret the source apportionment results.

Variability analysis is assessed following classical frequentist inference utilising a Monte-Carlo method (Helton, 1994). A succession of deterministic calculations is executed, each with different input values sampled from their respective distributions, to obtain probability distributions of the targeted outcomes.

The heterogeneity of each source is considered as a t-distribution for each property. The fingerprinting analysis of each sediment mixture is repeated by randomly sampling the source probability distributions. For the first iteration, the central value of the source distributions is used as a reference result. The corresponding output values are gathered to infer the probability distribution of the potential source contributions. Several samples must be collected for characterising each source to compute the mean and the SD of the analysed tracer properties.

196 **3. THE FINGERPRO PACKAGE**

Application of the functions in the package allows the user to i) characterise the different tracer properties and select the relevant variables; ii) unmix the sediment samples and quantify the different source apportionment; iii) assess the effect of the source variability; and iv) visualise and export the results. Thus, **FingerPro** package proposes a step by step procedure divided into three main sections to help users in their decisions.

203 **3.1** The example dataset

204 The package includes a soil dataset from a small Mediterranean catchment (4 km^2) that 205 contains high-quality radionuclide and geochemistry data to test the operation of the 206 functions and help the user to understand the model (Fig.1). This study area was selected 207 due to its heterogeneous land uses/land covers which are likely to exhibit large differences 208 in sediment tracer contents. Furthermore, the study area is located in a well-studied catchment where several studies of soil redistribution ¹³⁷Cs derived rates were pursued 209 210 (Quijano et al. 2016; Lizaga et al. 2018). Thus, soil redistribution rates were used to 211 evaluate **FingerPro** model as a suitable tool in the northern-central part of the Ebro basin. 212 The results obtained by Lizaga et al. (2018) found that net soil loss values were 4 times 213 higher in agricultural lands than in pine forest highlighting the importance of the 214 vegetation cover and land management to prevent erosion processes and subsequent land 215 degradation.

The study area dataset is composed of 21 source sediment samples from 4 different sources and 2 mixture samples. The sources are divided into agricultural (AG), old pine forest (PI), recent pine forest (PI1) and degraded soil named subsoil (SS) which occupy 9%, 32%, 58% and 1% of the catchment area, respectively. The agricultural land use is mainly composed of winter cereal crops and the pine afforestation forest is predominantly 221 *Pinus halepensis* Mill. The average temperature ranges from 5 °C to 18 °C, and the mean
222 annual rainfall is about 520 mm (AEMET).

3.2 *Input data*

The input variables need to be stored as an R table object. The dataset must satisfy the following requirements: i) the first column represents the sample *id*; ii) the second column is the source classification, containing target samples in the last place.

3.3 Characterising the sediment samples

One of the advantages of the **FingerPro** package is that it allows the user to analyse 228 and visually compare different tracer properties, using the state of the art of R packages: 229 230 The *boxPlot()* function displays a boxplot of each tracer property to help the user in the decision by visualising the different concentrations of each tracer versus the mixture 231 232 sample. A parameter (*columns*) with the number of tracer properties in the boxplots is 233 provided. The number of columns (ncol) refers to the number of plots per row in the 234 display (Fig.2). The boxPlot() function could be used for tracer selection by helping the 235 user to visualise and select the tracers based on the boxplots and its expert knowledge. 236 Thus, the user visualises in the example dataset that most of the ²¹⁰Pb_{ex} in the mixture sample likely comes from PI and PI1 sources and that ⁴⁰K is almost out of range (Fig.2). 237 238 Furthermore, by repeating this function after implementing each test for tracer selection, 239 users can envisage how representative the remaining tracers are.

The *correlationPlot()* function displays a correlation matrix of each tracer, divided by the different sources to help the user by testing the conservatism of tracers by visualising the relationships between the different tracers and sediment mixtures following the methodology proposed by Pulley et al. (2015). A parameter (*columns*) with the number of tracer properties in the correlation matrix is provided, along with the possibility to include the sediment mixture (mixtures = *T*) in the matrix or to exclude it (by default). In addition, in the correlation plot, once the users have selected the optimum set of tracers,
it is possible to visualise if the mixture samples fit inside the source distributions. If a
mixture sample is outside the sources distribution, then no solution exists or the mixing
model assumptions are not met.

The *PCAPlot()* function performs a principal components analysis on the given data matrix and displays a biplot of the results, divided by the different sources, to help the user in the decision. A parameter (components) with the number of principal components to display is included.

The LDAPlot() function performs a linear discriminant analysis and visualises the data in 254 255 the relevant dimensions. A parameter (P3D) allows the user to display a 3D LDA graph (Fig.3). This set of functions allows the user to visualise the principal components plot 256 257 and the linear discriminant plot after the statistical selection procedure. Thus, the plots 258 help the user to visually identify whether the excluded variables increase or decrease the discrimination capacity between sources. Furthermore, the LDAPlot() function was used 259 260 in the catchment example to visualise the number of sources that show good 261 discrimination with this set of tracers (Fig.3). The function shows a large overlap between 262 PI and PI1 that would suggest merging both sources. Thus, after grouping PI and PI1 the 263 discriminant plot shows better discrimination between the selected sources (Fig.3).

264 **3.4** Statistical test for selecting the optimal set of tracers

The selection of the optimal tracers is usually based on the two-step procedure proposed by Collins and Walling (2002), which includes some previous statistical procedures such as the "range test", the KW and the DFA test. Thus, **FingerPro** has included these functions to support user decisions. However, this procedure might remove too many tracers or include some inadequate properties and could, therefore, restrict the discrimination between sediment sources. Hence, the procedure is included as an individual and informative function to only use the steps needed and to prevent a reduction in the source discriminations. For this reason, the tracer selection procedure cannot be only based on statistical tests but also on the expert knowledge of the geomorphological and hydrological processes of the catchment (Blake et al. 2018). Thus, the boxplot chart, LDA plots and correlation plot included in the **FingerPro** package were implemented to help the users in the decision.

The *rangeTest()* function excludes the tracer properties of the mixture/s outside the lowest and highest values in the sediment sources.

The *KWTest()* function excludes tracers from the original dataset which do not show a significant difference between sources. This function performs a Kruskal-Wallis ranksum test using the kruskal.test() function from the R package **stats**. A parameter to select the p-value (*pvalue*) is provided.

283 The DFATest() function executes a stepwise forward variable selection, using the Wilk's Lambda criterion, which maximises the discrimination between the sources whilst 284 285 minimising the number of tracers. This function performs a stepwise forward variable 286 selection using the greedy.wilks() function from the R package klaR. A parameter to 287 select the niveau (niveau) for an approximate F-test decision is provided with a default 288 value of 0.1. This value could be reduced to be more restrictive in the tracer selection procedure. However, by reducing the value below 0.05 the statistical test could remove 289 290 the majority of the tracers with the subsequent decrease in the discrimination of the 291 different sources.

These three tracer selection methods were applied in the example dataset. In Fig.2 the tracers removed by each method can be seen and, based on the boxplot graph, to decide if it is suitable to use all of them or if the selected tracers represent a good approximation of the dataset. After the implementation of the range test function, we can see in the 296 boxplot graph that effectively Pb, Zn and Cr have been removed. However, there are other tracers such as ⁴⁰K, Sr, Fe, and V that remain in the dataset though they should not be 297 considered as tracers inside the source range. Furthermore, by using the LDA and PCA 298 299 plots we can decide if the use of other tracer selection methods decreases the discrimination or if by using them we could remove a tracer with specific information. 300 As shown in Fig.4, by removing ²²⁶Ra and Mn from the dataset by using the DFA after 301 302 the KW test, the LDA plot shows similar results. In addition, the arrows of the removed 303 tracers in the PCA plot were parallel to those that remain in the dataset. Thus, in this example, the plot information suggests that including or removing ²²⁶Ra and Mn should 304 305 not produce important variations in the discrimination of sources or the model results as is evident in Fig.4. 306

307 3.5 Sediment unmixing

The *unmix()* function assesses the relative contribution of the selected sediment sources for each mixture in the dataset. A parameter (*samples*) with the number of samples of the LHS is provided. The number of iterations (*iter*) in the source variability analysis is also configurable. However, if the number of iterations is set as 1, results are produced in a single analysis considering the sources mean value.

The *plotResults()* function displays a plot with the density distribution of the model solutions and a table with the mean value and the standard deviation of the model solutions (Fig.4). Besides, users can display the results in violin plots instead of density plots by adding the word *True* to the violin option.

After the tracer selection procedure, **FingerPro** results reveal that 18% of the mean sediment supply comes from agricultural land use and 34% and 47% from bare soil and pine forest, respectively. The small standard deviation of the three sources together with the high GOF value shows a good fit of the model to efficiently discriminate the selected sources (Fig.4). However, users should be cautious about using GOF as an assessment of
model reliability. Recent research has shown models with a high GOF can still deliver
inaccurate results (Palazón et al. 2015; Gaspar et al. 2019b), but also has shown that all
models with low GOF always deliver wrong results.

The results of the example dataset are supported by soil erosion rates estimated with 325 ¹³⁷Cs by Lizaga et al. (2018) in a Mediterranean catchment comprising the one studied 326 327 here. Thus, 18% of the sediment contribution is supplied from 9% of the area under 328 agricultural management and 47% of the contribution comes from pine forest that occupies 90% of the study catchment. Relatively speaking, the subsoil was the main 329 330 source with 34% of the contribution for only 1% of the area taken by the bare soil in the 331 study catchment. Our results highlight the hazards that subsoils have on supplying 332 important amounts of sediments to the water systems.

333 **3.6** Application in a Medium-Size catchment

In this section, as an example, the results of applying the **FingerPro** package in a 334 335 medium-size catchment (Lizaga et al. 2019) are described. Its larger surface area and a 336 higher number of sources result in a more complex unmixing. For this reason, all the tools added in the FingerPro package to help the users and characterise the unmixing dataset 337 338 are essential to reach robust results. Here, we highlight the most important decisions made 339 during the fingerprinting procedure and how the different tools included in the package 340 help the authors to unmix their data. To avoid repetition in this manuscript, only one 341 mixture sample collected at the outlet of the catchment is used to describe the FingerPro utilities. 342

Following the application of the range test and Kruskal Wallis test, the final selection was made based on expert judgment using the boxplots and correlation plots to finally identify the conservative tracers. Fig.5 illustrates how some tracers pass the selection 346 tests, such as RT, KW and DFA, but show non-conservative behaviour, i.e. LF, Fe, Ti 347 and Ca. In addition, if we analyse the correlation plot of the tracers that shows non-348 conservative behaviour, the sample mixture is located almost out of the point cloud. On 349 the other hand, the sample mixture is located inside the point cloud of the conservative tracers. Thus, based on this information it was decided to select the tracers after passing 350 351 the KW test using expert knowledge, thus obtaining more defined results and higher GOF 352 (Fig.6). Hence, all the tools added in the FingerPro package to remove the tracers that 353 violate the principles of conservativeness are needed in fingerprinting studies. This methodology suggests that including tracers with non-conservative or discordant 354 355 information into fingerprinting models does not add valuable information and could lead 356 the model to unpredictable results as it was found by Lizaga et al. (2020).

357 4. CONCLUSIONS

358 The application of mixing models it is necessary to understand source-tracer relationships what is generally performed by applying different software's to select the 359 360 best combination of sediment tracers. With FingerPro, diverse test and mechanisms have 361 been incorporated for tracer selection in a single software. Furthermore, the inclusion of 362 several plot functions such as *boxPlot*, *correlationPlot*, *LDAPlot* and *PCAPlot* allows the 363 user to check if the selected tracers are suitable for the unmixing process. This package for sediment source fingerprinting in hydrological systems offers a wider and easier 364 application in catchments affected by natural and human-induced changes. 365

Due to the increasing attention in tracing sediment methods and the need to select the best tracer combination, an open-source tool that includes all the steps for sediment unmixing is a key tool for the unmixing process. The example dataset included in **FingerPro** provides evidence of the large sediment supply and severe soil loss caused by land degradation and bare soil. In addition, the agreement between the unmixing results obtained from the example dataset with the ¹³⁷Cs derived rates supports the capability of the model for the sediment fingerprinting task. These results reflect the high importance of creating a low time-consuming and open-source unmixing model that combines the necessary tools to solve environmental issues such as reservoir siltation or soil loss and trace the sediment provenance.

FingerPro provides the users with tools to i) characterise the different sediment sources, establish correlations between the tracers and assist the selection of the optimal tracers, ii) graph the results using the state of the art of R packages iii) unmix sediment samples to estimate the apportionment of the sediment sources and iv) test the model using data from a Mediterranean study catchment included in the package.

In addition, the example dataset and the explained results of a medium-size catchment introduce the users on to the functioning and potential of the tools included in the **FingerPro** package while also showing the advantages of the fingerprinting technique to improve the understanding of sediment supply processes. Future research will concentrate on keeping the **FingerPro** package updated with new and upcoming fingerprinting techniques, such as the recently published Consensus Method (Lizaga et al., 2020) for improving the selection of tracers, avoiding non-conservative and dissenting tracers.

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- 401 Availability of data, material and Code availability:
- 402 The R package source files are available via:
- 403 GitHub platform: https://github.com/eead-csic-eesa/fingerPro
- 404 Name of code: fingerPro
- 405 Name of the manual: fingerPro_manual 1.2.pdf
- 406 Developer and contact address:
- 407 Ivan Lizaga Villuendas (lizaga.ivan10@gmail.com) / Borja Latorre
 408 (borja.latorre@csic.es)
- 409 Year first available: 2018
- 410 Software required: R Program language: R

411 Authors' contributions: All authors contributed to the study conception and design. Ivan Lizaga: Conceptualization, Writing - Original Draft, Software, Data Curation, 412 413 Methodology, Formal analysis, Investigation, Resources; Borja Latorre: 414 Conceptualization, Writing - review & editing, Software, Supervision, Methodology, 415 Formal analysis, Investigation, Resources; Leticia Gaspar: Conceptualization, Validation, Data Curation, Investigation, Resources, Writing - review & editing; Ana Navas: 416 417 Conceptualization, Methodology, Investigation, Writing - review & editing, Supervision, Project administration, Funding acquisition. 418

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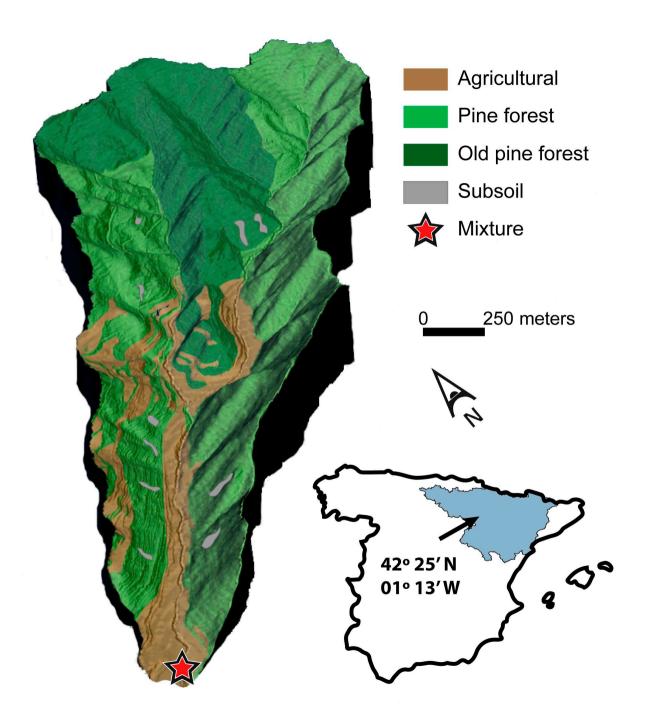
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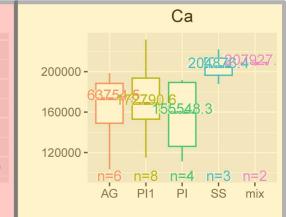
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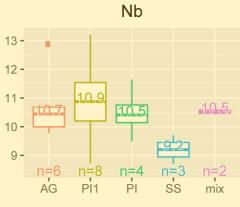
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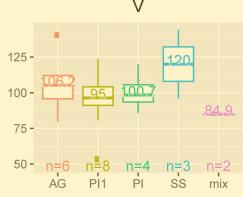
569 FIGURE CAPTIONS

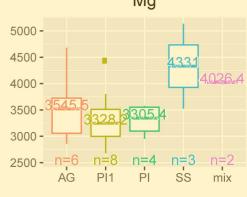
- Fig.1 Location of the study area. 3D picture of the catchment created with a DEM andland cover map
- 572 **Fig.2** Boxplot of the tracer properties included in the data example of a small 573 catchment. In different colours, the tracers removed by each statistical test
- 574 **Fig.3** LDA plot of the data example of a small catchment for the different land covers:
- agricultural (AG), old pine forest (PI); recent pine forest (PI) and subsoil (SS). a)
- 576 Before running the statistical test, the dataset shows collinearity. b & c) 2D and 3D
- 577 LDA display of the dataset after running the statistical selection. d) LDA display after
- 578 merging both pines sources PI and PI1
- 579 Fig.4 LDA, PCA and density plots of the unmixing process before and after the use
- of the DFA test for the different land covers: agricultural (AG), pine afforestation (PI)and subsoil (SS)
- **Fig.5** Correlation plots of seven of the tracer properties of the medium size catchment
- 583 example. Agricultural (AG), Forest (FO), subsoil (SS) and channel bank (CB)
- 584 **Fig.6** Scaled density plots and results of the unmixing process after the two different
- 585 tracer selection approaches for the different land covers: Agricultural (AG), Forest
- 586 (FO), subsoil (SS) and channel bank (CB)
- 587

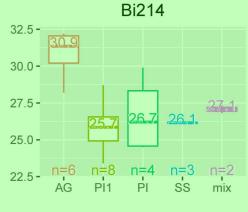


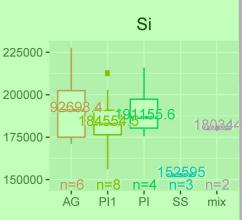












Zn 60 -50 -40 n=8 n=3 n=6 n=4 PI1 ÂĠ ΡI ŚŚ mix

Th232

в

n=4 Pl

Rb

29

n=4

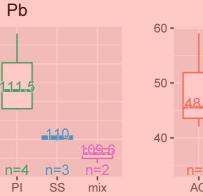
n=8 PI1

37.4

n<mark>=3</mark> SS

36

n=2



40 -

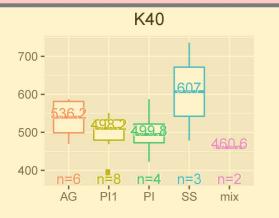
35.

30 -

25 -

n=6

AG



n=8

PI1

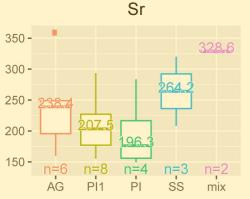
n=6

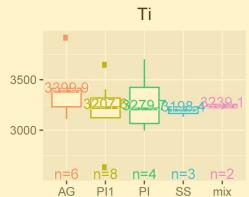
AG

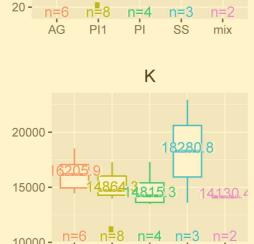
112 -

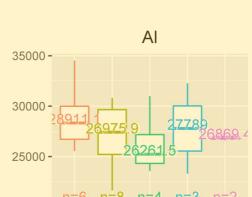
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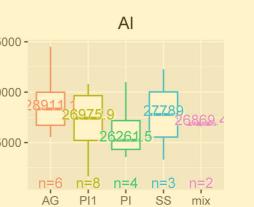
110-











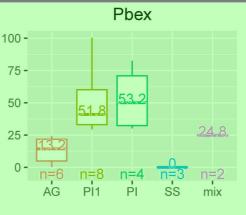
n=4

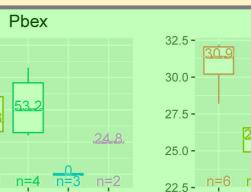
PI

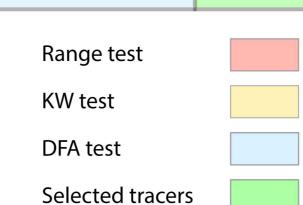
ŚŚ



PI1

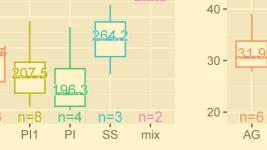






mix

ŚŚ



10000 n=6 n=8 n=4 n=3 n=2 U238

n<mark>=8</mark> Pl1

160 -

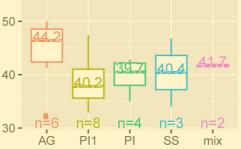
140 -

120

100 -

n=6

ÂG



Cr

n=4

ΡI

120

n=3

SS

mix

40 30.7 n<mark>=</mark>6 AG n=2 30 mix Fe 35000 -

n=6

AG

n=8

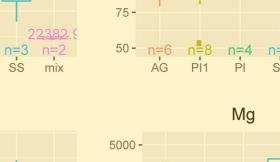
PI1

25000 -

30000 -

V







n=4

ΡI

Agriculture

Subsoil

____3

Old afforestation

New afforestation

Mixture sample

SS

n=2

mix

24 -

n<mark>=6</mark> AG

Land_Use

AG

PI1

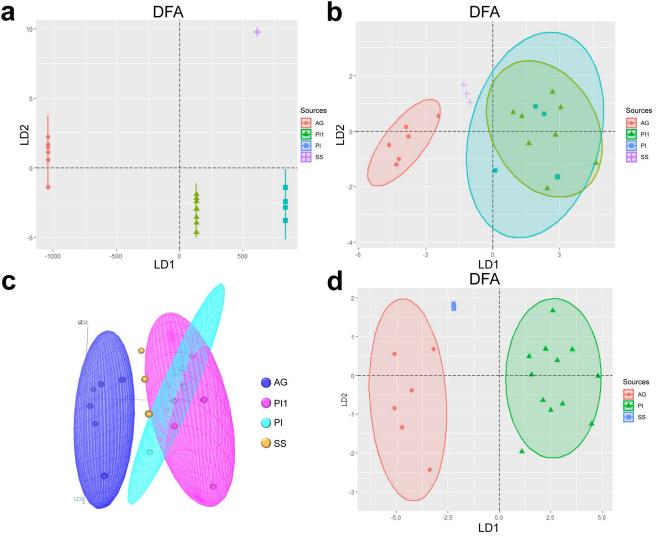
ΡI

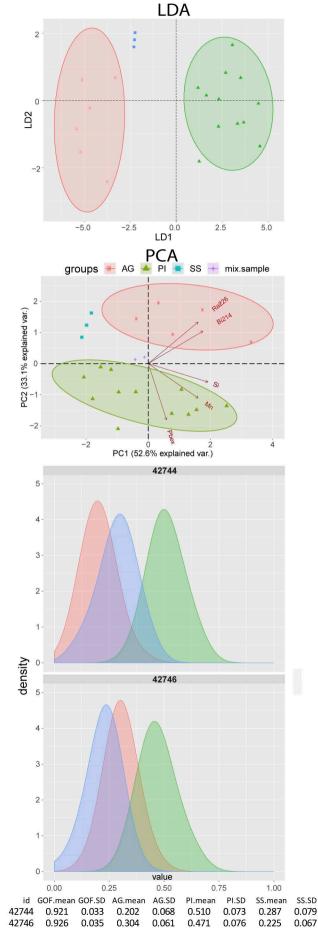
SS

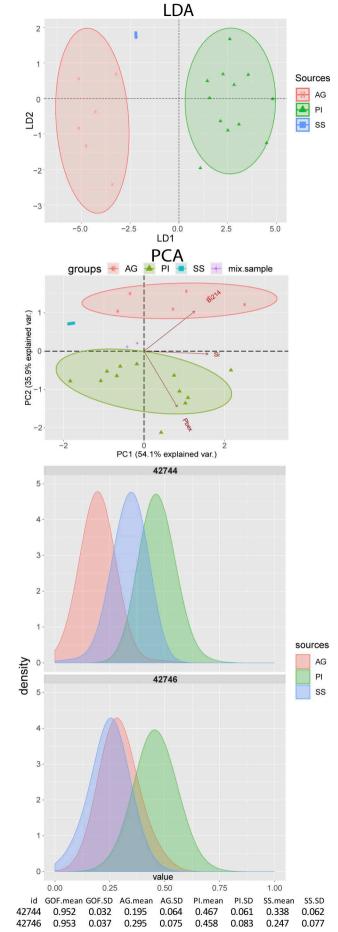
mix

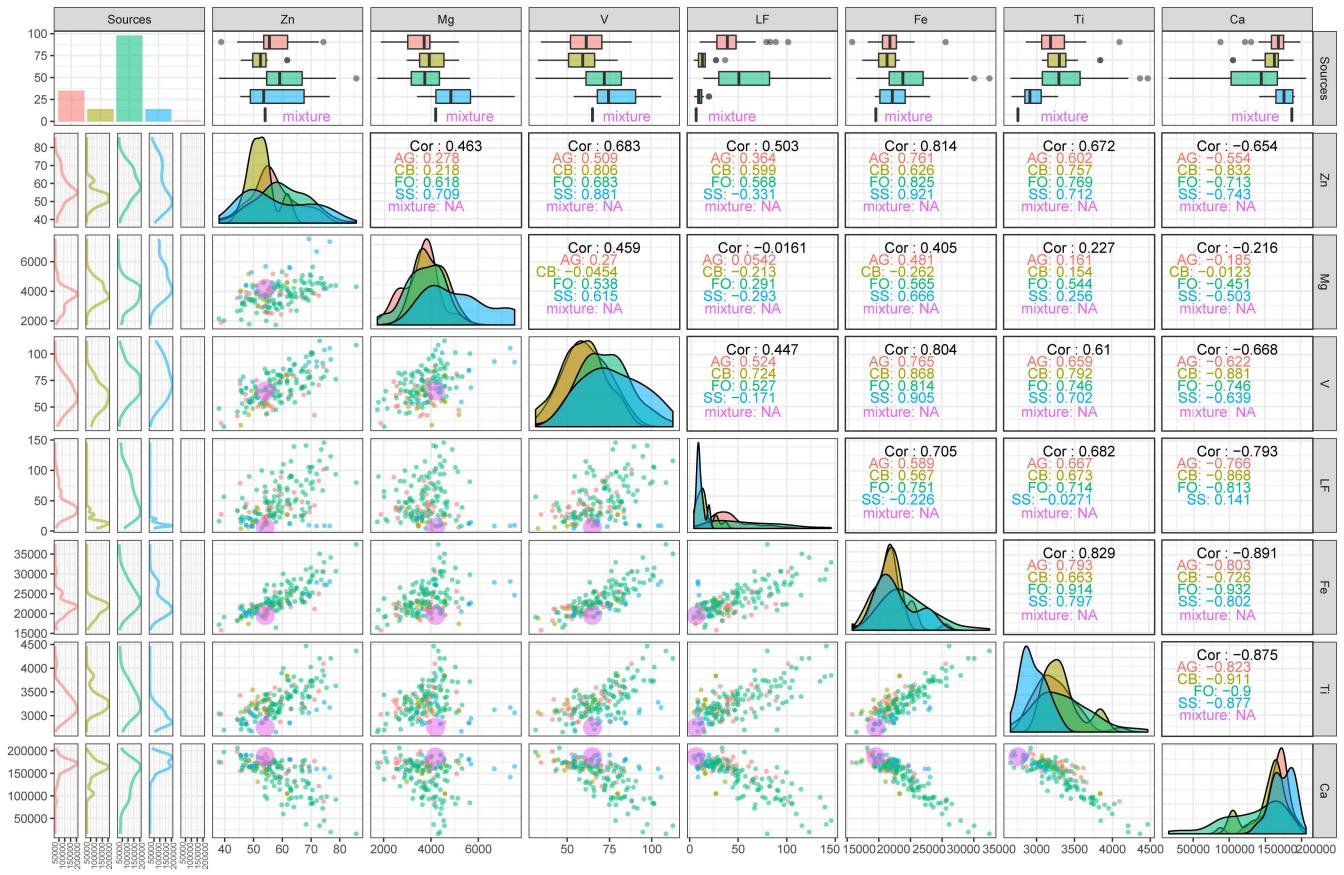
n=8

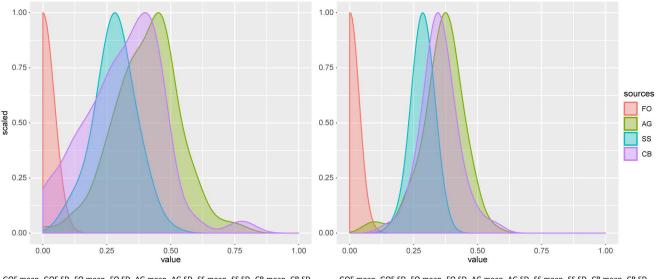
PI1











 GOF.mean
 GOF.SD
 FO.mean
 FO.SD
 AG.mean
 AG.SD
 SS.mean
 SS.SD
 CB.mean
 CB.SD

 0.713
 0.016
 0.002
 0.008
 0.403
 0.128
 0.280
 0.080
 0.315
 0.153

 GOF.mean
 GOF.SD
 FO.mean
 FO.SD
 AG.mean
 AG.SD
 SS.mean
 SS.SD
 CB.mean
 CB.SD

 0.933
 0.017
 0.003
 0.009
 0.362
 0.084
 0.285
 0.035
 0.351
 0.073