

1	Using ²¹⁰ Pb _{ex} measurements to quantify soil redistribution along two complex
2	toposequences in Mediterranean agroecosystems, northern Spain
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15	Abstract
16	Information on soil redistribution rates associated with the intricate patterns of
17	Mediterranean agroecosystems is a key requirement for assessing both soil degradation,
18	and off-site sediment problems that can affect downstream water bodies. Excess lead-
19	210 ($^{210}Pb_{ex}$) measurements provide a very effective means of documenting spatial
20	patterns of rates of soil redistribution in different landscapes, but to date the approach
21	has not been widely used in mountain Mediterranean landscapes. This research aims to
22	use 210 Pb _{ex} measurements to estimate soil redistribution rates on slopes uncultivated and
23	under cultivation, within two complex toposequences located in the vicinity of Estaña
24	Lake, characterized by an intricate mosaic of land use, steep slopes and anthropogenic
25	modification (e.g. terraces and tracks), which are typical of these agroecosystems in

26 northeastern Spain. A perceptual model is developed to account for the soil 27 redistribution dynamics along both toposequences. This provides a simple and novel 28 methodology adapted to Mediterranean agroecosystems, which besides using information on soil redistribution rates provided by ²¹⁰Pb_{ex} measurements, also takes 29 30 into account variations in land use and the presence of linear landscape elements, which 31 modify runoff and soil redistribution processes and sediment connectivity along the 32 toposequences. The results show that erosion predominated on the steep cultivated 33 slopes, but lower soil redistribution rates were found on the uncultivated slopes. On the 34 flat areas at the bottom of both transects, deposition was dominant. Variations in land 35 use and the presence of linear landscape elements control soil redistribution processes. 36 Such elements can perform the role of Ecological Focus Areas (EFAs), proposed within 37 'The Green' Common Agricultural Policy for 2014, in which at least 7 % of a farmer's 38 land should comprise EFAs, which can include terraces, landscape features, buffer strips 39 and afforested areas.

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41 Keywords: ²¹⁰Pb_{ex}; Soil erosion; Soil redistribution rates; land use; CAP; linear
42 landscape elements; Mediterranean agroecosystems.

43

44 **1 Introduction**

Soil degradation by water erosion represents one of the major environmental problems facing the sustainable management of soil and soil productivity. Cultivation is seen as a key factor promoting soil mobilization and soil loss. Other related effects, including the mobilization and transport of sediment-associated contaminants (pesticides, fertilizers) and the siltation of wetland areas must also be taken into account to protect fragile agroecosystems. In addition, soil erosion transfers soil organic carbon from topsoil to deposition sinks in the landscape and promotes soil carbon replacement at eroded sites
(Ritchie and McCarty, 2003).

53 Increased awareness of the problems of soil loss in the last decade has promoted actions 54 to conserve soil under the European Common Agricultural Policy (CAP), including the 55 most recent Green Areas initiative. In Mediterranean mountain agroecosystems large 56 areas of agricultural land were abandoned during the past century as a result of major 57 socio-economic changes. In recent years, however, some steep marginal lands have 58 been returned to cultivation under the European Agrarian Policy (García-Ruiz et al., 59 2008; García-Ruiz, 2010; Gaspar et al., 2013). The study area selected for this research 60 is a good example of mountain areas in northern Spain, which illustrates many of the 61 problems associated with steep slopes, high rainfall intensity, changes of land use, and 62 especially the abandonment of the less productive land located on steep slopes. Previous 63 studies highlight the importance of soil erosion in the study area, especially in cultivated areas. For cropland areas, Navas et al. (2012a) used caesium-137 (¹³⁷Cs) measurements 64 to estimate erosion rates as high as 108 Mg ha⁻¹ year⁻¹, while López-Vicente and Navas 65 (2010) predicted severe erosion rates (> 100 Mg ha⁻¹ year⁻¹), using a combination of the 66 67 RMMD and SED models, in gullies on Keuper facies. In the study area, the term 68 'uncultivated' has been used to refer to a range of conditions including areas of 69 undisturbed natural vegetation and areas under Mediterranean forest and scrub, as well 70 as old abandoned fields in long-term fallow (> 100 years), which are now covered by 71 dense scrub, and the more recently abandoned fields (ca. 50 years) that have a much 72 reduced vegetation cover. This leads to the development of a spatial pattern of vegetated 73 patches and bare cultivated inter-patch areas, affecting water redistribution and it is well 74 established that vegetation development and vegetation structure affect the connectivity 75 of runoff and soil redistribution processes on slopes (Cerdá 1997).

Lead-210 (²¹⁰Pb) is a natural geogenic radioisotope (half life, 22.2 yr) of the uranium 76 decay series. Decay of radium-226 in the soil and regolith releases radon-222 (²²²Rn), 77 which in turn decays to ²¹⁰Pb. Some of the ²²²Rn diffuses upward through the soil and 78 enters the atmosphere where it decays to ²¹⁰Pb and is returned to the earth's surface as 79 fallout. Fallout ²¹⁰Pb reaching the soil surface is rapidly adsorbed by clay minerals and 80 81 organic matter, and its subsequent redistribution is controlled by soil redistribution processes in a manner similar to ¹³⁷Cs (Walling and He, 1999a, b). This fallout ²¹⁰Pb is 82 termed unsupported or excess 210 Pb (210 Pb_{ex}), since it is not in equilibrium with its 83 parent ²²⁶Ra. ²¹⁰Pb_{ex}, like ¹³⁷Cs, offers the potential for use as a tracer in estimating rates 84 of soil redistribution. However, ²¹⁰Pbex measurements have been less widely used for 85 estimating soil redistribution rates than ¹³⁷Cs, although their use has increased 86 87 significantly in recent years (e.g. Wallbrink and Murray, 1993; Walling and Quine, 88 1995; He, Q. and Walling, D.E. 1996; Zhang, et al., 2006; Kato et al., 2010; Porto and 89 Walling, 2012; Benmansour et al., 2013).

The continuous input of ²¹⁰Pb_{ex} fallout through time means that the contemporary 90 91 ²¹⁰Pb_{ex} inventory in the soil will reflect soil redistribution and thus loss and gain of 92 210 Pb_{ex} occurring within a period equivalent to four times the half-life, and thus the past 100 years (Walling et al., 2003). However, the effect of past changes in the ²¹⁰Pb_{ex} 93 94 inventory, caused by erosion and deposition, on the contemporary inventory, will 95 progressively decline as the period of time elapsed increases and this must be taken into 96 account when interpreting the impact of the erosional history of a study site on the magnitude of the contemporary ²¹⁰Pb_{ex} inventory. This inventory will clearly be more 97 98 sensitive to recent soil redistribution, and the estimate of the mean rate of soil 99 redistribution for the past ca. 100 years provided by the conversion model used to 100 estimate the soil redistribution rate from a comparison of the inventory measured at a

101 sampling point with the local reference inventory is likely to be biased towards the102 recent erosional history of the study site.

103 In order to understand soil redistribution dynamics in the intricate toposequences that 104 are characteristic of the typical agroecosystems of northern Spain, it is important to 105 know how the interfacing of patches of different land use and linear landscape elements 106 modify soil redistribution processes and the sediment connectivity along the slopes. The use of ²¹⁰Pb_{ex} measurements provides a means of investigating such systems. Their use 107 108 to investigate both cultivated and uncultivated soils and to quantify sediment sources 109 and sinks along slopes of different aspect represents a novel application, particularly within a mountain agricultural area. The use of ²¹⁰Pb_{ex} measurements to document soil 110 111 redistribution rates and analysis of the factors that affect soil redistribution along 112 toposequences of different aspect affords a means of developing an improved 113 understanding of the role of land use, soil type and slope gradient in Mediterranean 114 agroecosystems. Additionally, the development of a perceptual model of soil movement 115 of redistribution rates, which take into account changes in land uses and the presence of 116 linear landscape elements, is seen as potentially offering a new tool to elucidate 117 sediment connectivity in intricate landscapes. This is of importance for developing 118 'green' agricultural practices, as 'The Green' CAP proposes that at least 7 % of farmland 119 should be converted to Ecological Focus Areas.

The objectives of this study were therefore to use ${}^{210}Pb_{ex}$ measurements to estimate the long-term mean annual rate of soil redistribution on cultivated and uncultivated soils along two slope transects representatives of Mediterranean agroecosystems in NE of Spain. Its results aim to contribute to a better understanding of the impact of land use and the presence of linear landscape elements (both natural features and anthropogenic infrastructure) on soil redistribution processes along toposequences. Additionally,

126 assessment of the importance of natural features for trapping and storing eroded soil, as 127 promoted by the new Common Agricultural Policy (CAP) is a key requirement for both 128 the sustainable management of the soil resource and the protection of downstream 129 aquatic ecosystems from degradation resulting from increased sediment loads. Finally, 130 the development of a perceptual model of soil movement aims to elucidate how linear 131 landscape elements contribute to patterns of soil redistribution along cultivated and 132 uncultivated toposequences.

133

134 2 Material and methods

135 2.1 Study area

136 The study was conducted along two representative toposequences located in the Spanish 137 central Pre-Pyrenees (NE Spain), close to the northern boundary of the Ebro river basin 138 (Figure 1). This area includes a freshwater lake. Estaña Lake, in the lower part of the 139 landscape that has been under regional protection since 1997 and is included in the 140 European NATURA 2000 network as a Site of Community Importance. The average 141 annual precipitation is 595 mm (1997-2006) with two wet periods, spring and autumn, 142 and a dry summer with high intensity rainfall events extending from July to October. 143 The average annual temperature is 12.2° C, with thermal inversions common during the 144 winter (López-Vicente et al., 2008). The Mediterranean agroecosystem of the study area 145 comprises an intricate landscape, characterized by abrupt relief with slope gradients up 146 to 34 %. The cultivated and uncultivated areas are heterogeneously distributed. The 147 cultivated fields are located in the lower and mid slope areas and are separated by 148 vegetation strips, while uncultivated areas predominate on the steep slopes. Winter 149 barley is the main crop and, as indicated above, uncultivated areas include areas of 150 Mediterranean forest, scrubland and abandoned fields recolonised by natural vegetation.

151 The predominant soil types along the toposequences are stony Calcisols and Regosols. 152 Leptosols are restricted to the upper part of the slope under Mediterranean forest 153 underlain by Muschelkalk facies, and Gypsisols cultivated for cereals are restricted to 154 the lower part of one of the transects underlain by Keuper facies.

155 Two representative hillslope transects, extending from the divide to the lake, were 156 selected to represent different toposequences within this agroecosystem (Figure 1). A 157 total of 34 sampling sites, approximately 50 m apart were established along both 158 transects. However, it was recognised that tillage erosion in fields delimited by furrows 159 and tracks can cause significant soil redistribution both at the head and the bottom of the 160 fields (Gaspar, 2011), and thus the spacing of the sampling sites on the lower cultivated 161 part of the ST was reduced to 25 m, in order to provide a reliable representation of soil 162 redistribution in this area (Figure 1).

163 The northern transect NT (S-N) is 300 m long and characterized by a 10 % slope. Its 164 altitude ranges from 711 to 682 m and the seven sampling sites are located on a gentle 165 north facing slope occupied exclusively by uncultivated areas. Regosols are restricted to 166 the upper part of the transect while Calcisols predominated in the rest of the transect. 167 The southern transect ST (N-S) is 1110 m long and extends down a steeper south facing 168 slope (21 % slope) with an altitude ranges from 894 to 676 m. The transect crosses 169 patches of different land use. The uncultivated areas are located primarily on the upper 170 and midslope sections of the transect on Calcisols and Regosols, whereas the cultivated 171 fields are located on the midslope on stony Calcisols and Regosols and on the bottom 172 slope on Gypsisols with a low stone content. Leptosols are found within the upper part 173 of the transect under Mediterranean forest and on a thick Muschelkalk outcrop on the 174 midslope. ST includes 27 sampling sites and is characterized by a rugged topography 175 and the presence of agricultural terraces, a thick Muschelkalk outcrop on the midslope and vegetation strips, which have an important effect on hydrological processes as theyreduce the local slope gradient, intercept runoff and trap eroded sediment.

178 Nine bulk cores were collected from sampling points located on cultivated soils and 24 179 sectioned profiles were collected from the sampling points on uncultivated soils, which 180 were also the subject of another study investigating the depth distribution of 181 unsupported ²¹⁰Pb (Gaspar, 2011). Sampling site ST-14 is located on a thick Muschelkalk outcrop and soil samples for ²¹⁰Pb_{ex} measurements were not collected from 182 183 this point. The bulk cores obtained from the cultivated areas were collected using a 8.0 184 cm diameter hand-operated core sampler. The core depth always exceeded the plough 185 depth (ca. 20 cm), with a maximum of 55 cm. The sectioned profiles were collected 186 using a 10 x 10 cm steel box corer (Navas et al., 2008) at 2 cm depth intervals to a 187 maximum depth of 10 - 14 cm depth, which had been shown by previous work in the study area to include the complete ²¹⁰Pb_{ex} profile in uncultivated soil (Gaspar, 2011). At 188 189 each uncultivated sampling site, a three-sided frame was driven into the ground with the 190 open end of the sampling frame facing downslope. The soil downslope of the sampler 191 was carefully removed until a block of soil was enclosed within the sample frame. A 192 blade was inserted into a series of grooves spaced at 2 cm on the sides of the device to 193 section the profiles.

The samples collected from each sampling point along the transects were dried, gently disaggregated and sieved to < 2mm. The stone content (%) was determined as the proportion > 2mm. The < 2mm fraction was analysed to obtain the total 210 Pb_{ex} inventory (Bq m⁻²), the soil organic carbon (SOC) content, and the mean clay, silt and sand content. The SOC content was determined by the dry combustion method using a LECO RC-612 multiphase carbon analyzer. In this case, a sub-sample of the < 2 mm fraction is inserted into a quartz tube, heated to 550 °C and the SOC is oxidized to CO₂, 201 which is selectively detected by an infrared (IR) gas analyser. Grain size analysis of the 202 < 2mm fraction to determine the sand, silt and clay content (%) was undertaken using a 203 laser granulometer. Prior to grain size analysis, organic matter was removed from the 204 samples using 10% H₂O₂ heated to 80 °C and the mineral sediment was ultrasonically 205 dispersed.

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2.2 Using ${}^{210}Pb_{ex}$ as a sediment tracer

To determine the ²¹⁰Pb_{ex} inventory at each sampling point, a representative aliquot of 208 209 the < 2mm fraction of the bulk core or the individual sections of the sectioned cores was 210 placed into a cylindrical plastic container and sealed for 40 days prior to assay in order to achieve equilibrium between ²²⁶Ra and its daughter ²¹⁴Pb. The ²¹⁰Pb_{ex} activity in the 211 212 sample was measured by gamma-ray spectrometry, using a high resolution low energy 213 coaxial HPGe detector coupled to an amplifier (broad energy detector (BeGe)). The 214 detector had an efficiency of 30 % and a resolution of 1.9 keV, and was contained 215 within a lead shield to reduce the background. Calibration was achieved using standard 216 certified samples with the same geometry and bulk density as the measured samples. 217 Count time was typically ca. 86,400 s, providing results with an analytical precision of ca. 10 - 15 % at the 95 % level of confidence. The total ²¹⁰Pb activity in the samples was 218 measured at 46.5 keV, and the ²²⁶Ra activity was obtained by measuring the activity of 219 ²¹⁴Pb, a short-lived daughter of ²²⁶Ra at 351.9 keV. The detection limits in Bq kg⁻¹ for 220 ²¹⁰Pb and ²¹⁴Pb were 7.45 and 1.26 Bg kg⁻¹, respectively. The ²¹⁰Pb_{ex} activity was 221 determined by subtracting the ²²⁶Ra activity from the total ²¹⁰Pb activity. The ²¹⁰Pb_{ex} 222 inventories for individual sampling points were calculated using the measured ²¹⁰Pb_{ex} 223 224 activities. With the sectioned cores this involved summing the values for the individual 225 sections.

Estimates of soil redistribution rates are derived from ²¹⁰Pbex measurements by 226 227 comparing the total inventory for an individual sampling soil with the local reference 228 inventory for the study area and using a conversion model to estimate the erosion rate 229 represented by a reduced inventory or the deposition rate represented by an increased inventory. In order to establish the ²¹⁰Pb_{ex} reference inventory, two sectioned profiles 230 231 and seven bulk cores were collected from an undisturbed location adjacent to the 232 sampled transects, with minimal slope and no evidence of erosion or deposition, such 233 that no sediment redistribution was likely to have occurred over the past 100 years. The undisturbed nature of the reference sites was confirmed by the ²¹⁰Pb_{ex} depth profiles that 234 235 provided a well-defined exponential depth distribution. Estimates of soil redistribution 236 rates along the two transects investigated were obtained using the conversions models 237 for cultivated and uncultivated soils described by Walling and He (1999a) and Walling 238 et al. (2011).

239 Soil redistribution rates on cultivated soils were estimated using a mass balance model 240 (mass balance model 2) developed at the University of Exeter (see Walling and He, 1999a). The model takes into account the continuous atmospheric deposition of ²¹⁰Pb_{ex} 241 242 and its subsequent decay and its redistribution in association with soil erosion and 243 deposition. In addition, the model considers the effect of particle size selectivity of 244 sediment mobilization, and the transport and the removal of freshly deposited fallout ²¹⁰Pb_{ex} by erosion, before its incorporation into the tillage horizon. The basic form to 245 estimate the erosion rate R (kg m⁻² year⁻¹) can be expressed as Equation 1: 246

247
$$\frac{dA(t)}{dt} = (1 - \Gamma)I(t) - \left(\lambda + P\frac{R}{d}\right)A(t)$$
(1)

where A(t) is the cumulative ²¹⁰Pb_{ex} inventory (Bq m⁻²); *d* the tillage depth (kg m-2); λ is the ²¹⁰Pb decay constant (year⁻¹); I(t) the annual fallout ²¹⁰Pb_{ex} deposition flux at time *t* (Bq m⁻² year⁻¹); Γ the proportion of the freshly deposited ²¹⁰Pb_{ex} fallout input removed by water erosion before incorporation into the tillage layer; and *P* the particle size correction factor to take account of differences between the grain size composition of the mobilised sediment and the original soil (Walling ad He, 199a; Walling et al., 2003). The model used to estimate the deposition rate R'(kg m⁻² year⁻¹) takes the form indicated by Equation 2:

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$$A_{c,ex} = \int_{t_0}^{t} R' C_d(t') e^{e^{-\lambda(t-t')}} dt'$$
(2)

where $A_{c, ex}$ is the ²¹⁰Pb_{ex} inventory (Bq m⁻²) and $C_d(t')$ represents the concentration of ²¹⁰Pb_{ex} in deposited sediment (Bq kg⁻¹). $C_d(t')$ can be estimated as the weighted mean ²¹⁰Pb_{ex} activity of the sediment eroded from the upslope contributing area.

A modified version of the diffusion and migration conversion model developed for ^{137}Cs measurements (Walling and He, 1999b) was used to estimate soil redistribution rates from $^{210}Pb_{ex}$ inventories at the uncultivated sampling points. This model assumes a constant fallout of $^{210}Pb_{ex}$ and takes into account post-depositional redistribution processes and their influence on the $^{210}Pb_{ex}$ depth distribution (Walling et al., 2011).

A diffusion coefficient D (kg² m⁻⁴ year⁻¹) is used to represent the net effect of the slow vertical redistribution of ²¹⁰Pb_{ex} by physicochemical and biological processes. The rate of soil loss R (kg m⁻² year⁻¹) can be estimated from the reduction of the ²¹⁰Pb_{ex} inventory at the sampling point, relative to the reference inventory for the study site ($A_{u,ls}(t)$) and a model-derived estimate of the ²¹⁰Pb_{ex} content of the surface soil ($C_u(t')$), as indicated by Equation 3:

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$$\int_{0}^{t} PRC_{u}(t')e^{-\lambda(t-t')}dt' = A_{u,ls}(t)$$
 (3)

The deposition rates R' (kg m⁻² year⁻¹) can be estimated (Equation 4) from the increase in the ²¹⁰Pb_{ex} inventory compared with the local reference value ($A_{u,ex}(t)$), and the ²¹⁰Pb_{ex} content of deposited sediment soil ($C_d(t')$) (Walling et al., 2011).

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$$R' = \frac{A_{u,ex}}{\int_0^t C_d(t') e^{-\lambda(t-t')} dt'}$$
(4)

276 $C_d(t')$ can be estimated as the weighted mean ²¹⁰Pb_{ex} activity of the sediment 277 eroded from the upslope contributing area.

eroded from the upslope contributing area.
When applying the models to the ²¹⁰Pb_{ex} measurements obtained from the two transects,
values of 4 kg m⁻² and 1.0 were assumed for the relaxation depth (*H*) and particle size

correction (*P*) parameters, respectively, in both models and a value of 1.0 was assumed
for the proportion parameter (*y*) in the mass balance model.

282 Once estimates of the soil redistribution rate were derived for each of the sampling 283 points, the methodology proposed by Collins et al., (2001) was applied to each 284 toposequence (NT and ST) to estimate the net soil redistribution rate associated with the 285 two transects. In addition, in order to refine and adapt this technique for application to 286 intricate Mediterranean landscapes, it is important to take account of the effects of linear 287 landscape elements. The presence of agricultural terraces, buffer strips, rock outcrops 288 and tracks can have an important effect on downslope runoff and sediment transfer as 289 they reduce the slope gradient and length, and trap the eroded soil, influencing the 290 distribution of areas of erosion and deposition along the slope.

Statistical analysis was performed by one-way analysis of variance (ANOVA), and the means were subjected to a least-significant difference test (F test) to indicate the main differences in 210 Pb_{ex} inventories and soil properties between cultivated and uncultivated sites, and the differences in the soil redistribution rates estimated from the 210 Pb_{ex} measurements between the different land uses, soil types and slope gradient.

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3 Results and discussion

298 3.1 Assessment of soil redistribution rates using $^{210}Pb_{ex}$ inventories

The reference ²¹⁰Pb_{ex} inventory for the study area estimated from nine sampling points 299 300 located on undisturbed soil with minimal slope adjacent to the study transects is 2019.8 \pm 215.8 Bq m⁻². Figure 2 shows the depth distributions of ²¹⁰Pb_{ex} for a representative 301 302 reference soil profile in the study area. This reference inventory is very similar to that 303 reported for the area from a preliminary study reported by Gaspar et al. (2013) and is within the range of ²¹⁰Pb_{ex} reference inventories reported by Sanchez-Cabeza et al. 304 (2007) for different parts of northern Spain (between 1044 and 8204 Bq m⁻², depending 305 306 on the mean annual rainfall of the study site). However, the reference inventory 307 obtained for the study area is smaller than values reported by other authors for different areas of the world, for example: 5170 Bq m⁻² in the UK (Walling and He, 1999a), 5730 308 and 12860 Bq m⁻² in China (Zhang, et al., 2003 and Zhang, et al., 2006, respectively), 309 6310 Bq m⁻² (Kato et al., 2010) and 19703 Bq m⁻² (Wakiyama et al., 2010) in Japan, 310 5266 (Porto et al., 2006), 14572 Bg m⁻² (Porto et al., 2009) and 7598 Bg m⁻² (Porto and 311 312 Walling, 2012) in Italy, 34000 in Taiwan (Huh and Su, 2004), and between 3580 and 10060 Bq m⁻² for different floodplain sites in England and Wales (Du and Walling, 313 314 2012).

The ²¹⁰Pb_{ex} inventories recorded along the two study transects showed significant variability, reaching a maximum of 7298.2 Bq m⁻² (Table 1). The ANOVA test indicated that the ²¹⁰Pb_{ex} inventories were higher for cultivated soils than for uncultivated soils, although the differences were not statistically significant. A similar pattern has been previously documented in the study area for ¹³⁷Cs, and this trend confirms the importance of land use in controlling fallout radionuclide inventories and thus soil redistribution rates in the local area (Navas et al., 2012a; Gaspar et al., 2013).

322 The main soil properties analyzed showed values consistent with the characteristics of 323 Mediterranean agroecosystems. Information on stone content, SOC content and grain size composition for cultivated and uncultivated soils are presented in Table 1. The SOC and stone content were significantly higher in uncultivated soils. In cultivated soils the maximum values of SOC did not exceed 2.3 %, confirming the impact of long-term and intense agricultural use on SOC content (Navas et al., 2011). The relative magnitude of the clay, silt and sand fractions varied greatly between the sampling points. However, no significant difference was found between the two land uses, with silt-loam being the predominant texture.

After isolating the land use factor, only SOC showed significant differences between different soil types. For cultivated soils, higher inventories of 210 Pb_{ex} were found on Gypsisols, while Calcisols showed significantly higher mean values of SOC and slightly higher mean values of stone and sand content. In uncultivated areas, significantly higher mean values of SOC was found on Leptosols, which in turn have slightly higher values of 210 Pb_{ex} inventory and stone content (Table 2).

337 For the northern transect (NT), the lack of significant reduction or increase in inventory 338 values for the sampling points indicates that these points have not experienced 339 significant soil redistribution over the past 100 years, and particularly in recent years. In 340 contrast, significant increases and reductions in inventory values, relative to the 341 reference inventory, for the sampling points on the southern transect (ST), particularly 342 for cultivated points, suggest that these points have experienced appreciable soil loss or 343 deposition over that period, and indicate that significant soil redistribution has occurred 344 along transect ST, in marked contrast with the NT transect.

The soil redistribution rates (Mg ha⁻¹ year⁻¹), derived from the ²¹⁰Pb_{ex} inventories using the models described above and shown in Figure 3, indicate that erosion rates range between 0.1 and 83.7 Mg ha⁻¹ year⁻¹ and sedimentation rates range between 0.08 and 74.8 Mg ha⁻¹ year⁻¹. The highest values were found on cultivated soils, whereas on

uncultivated soils, erosion and deposition rates did not exceed 2.4 and 5.6 Mg ha⁻¹ 349 year⁻¹, respectively (Table 3). As shown in Figure 3, the soil redistribution rates follow 350 quite closely the changes in land use. For the uncultivated transect (NT) most sampling 351 points recorded low erosion rates, with a maximum of 2.4 Mg ha⁻¹ year⁻¹ (NT-6) and 352 353 most values close to stability (NT-1, NT-4, NT-5). The highest sedimentation rates were found at the bottom part of the transect (NT-7) and did not exceed 5.6 Mg ha⁻¹ year⁻¹. 354 355 On the contrary, the combined effects of topography and tillage have caused different 356 patterns of soil redistribution along the ST. Uncultivated areas on ST evidence similar 357 soil redistribution rates to those found on NT and soil stability predominates. In the 358 upper part of ST the dense forest protected the soil surface from erosion (ST-1, ST-3) and higher deposition rates, that did not exceed 1.3 Mg ha^{-1} year⁻¹ (ST-20), were 359 identified on the relatively flat areas (ST-6, ST-10, ST-17). The highest erosion rate 360 within the uncultivated areas was located at ST-19 (2.3 Mg ha⁻¹ year⁻¹), which 361 362 corresponds to open scrubland. On the steeper cultivated slopes, sampling points ST-13, ST-15 and ST-16 recorded high erosion rates (between 5.4 and 54.20 Mg ha^{-1} year⁻¹). 363 364 In contrast, on cultivated flat areas at the bottom part of the ST, sampling points ST-23, 365 ST-24, ST-26 and ST-27 evidenced the highest deposition rates, but the highest erosion rate was also found at ST-25 (83.7 Mg ha⁻¹ year⁻¹). Previous research in this area with 366 ¹³⁷Cs and ²¹⁰Pb_{ex} (Gaspar et al., 2013) provide evidence that tillage erosion was 367 368 important in these cultivated fields. The soil redistribution rates estimated from the 210 Pb_{ex} measurements are in agreement with those obtained from 137 Cs measurements in 369 370 the same study area, using appropriate conversion models (Soto and Navas, 2004, 2008), which ranged between 2.6 and 31.9 Mg ha⁻¹ year⁻¹ for erosion rates, and 371 between 0.2 and 24.5 Mg ha⁻¹ year⁻¹ for deposition rates. These results demonstrate the 372 373 important effect of agricultural activities on soil redistribution. The presence of ridges and furrows causes a local increase in slope gradient on the side of the furrow, relativeto the natural slope, which will increase rates of interrill erosion (Junge et al., 2010).

376 The mean erosion rates for the cultivated fields were significantly higher than for the 377 uncultivated areas. Slightly higher erosion rates were found on Regosols than on 378 Leptosols and Calcisols, although differences were not significant, while on Gypsisols 379 the mean erosion rates were significantly higher. Table 4 indicates that erosion rates 380 were similar in areas with average slope between 0 to 12 % and 12 to 24 % and that 381 these rates appeared to be higher than those on steeper slopes (> 24 %), although the 382 difference was not statistically significant. The mean deposition rates were significantly 383 higher for cultivated areas and on Gypsisols. However, unlike the erosion rates, 384 significantly higher deposition rates were found on flat areas (0 to 12 %) (Table 4).

385 In the study area, land use, soil type and slope gradient are linked. Most of the 386 uncultivated profiles were on Leptosols and Calcisols, located along the upper part of 387 ST and along NT and these points had the lowest rates of soil redistribution. While, 388 most cultivated sampling points were on Gypsisols, these were located on the flatter 389 lower part of ST, which recorded the highest redistribution rates (both erosion and 390 deposition). On Regosols, the cultivated sampling points were on steep slopes, while the 391 sampling points on uncultivated areas consisted of open scrubland. Both favoured 392 redistribution processes.

Principal components loadings and biplot after varimax rotation (Table 5, Figure 4) show that for erosion rates, three components were retained with eigenvalues higher than one, explaining 74 % of total variance. The first principal component, which represents 32 % of the total of variance, showed high values for the variables related to grain size. The second component, with 29 % of total variance, showed high loading values of the parameters related to land use and erosion rates, which were negatively correlated with SOC and also

399 with stone content but the estimated communality of this particular variable is lower than 400 0.5 and represents a low proportion of the variance. The third component was associated 401 with the slope factor, which was negatively correlated with SOC, and represents 13 % of 402 variance (Table 5.a). For deposition rates, two components explained 78 % of the total 403 variance. The first component, which represents 54 % of the total variance, showed high 404 loading values for the parameters related to land use and deposition rates, which were 405 negatively correlated with SOC, slope factor and stoniness, while the second component, 406 with 24 % of total variance, showed high loading values of the parameters related to grain 407 size (Table 5.b).

408 Although PCA cannot be used numerically for prediction purposes, the PCA biplot (Figure 409 4a, 4b) is of interest as it indicates the level of correlation between the analyzed variables. 410 Both erosion and deposition rates are positively correlated with land use. Likewise, the fact 411 that SOC is negatively correlated with erosion rates can be interpreted to mean that soil 412 loss is associated with loss of organic carbon (Figure 4.a), as reported for similar 413 environments by Navas et al. (2012b) and in agreement with Ritchie and McCarty 414 (2008), who also reported strong links between soil redistribution and soil organic 415 carbon concentrations in agricultural soils. In addition, the fact that stone content and 416 slope are negatively correlated with deposition rates can be interpreted to mean that in flat 417 areas evidencing a lower stone content deposition processes predominate (Figure 4.b).

Despite the small sample size (n=20 for erosion rates and n=13 for deposition rates), the communality of most variables was higher than 0.5 (Table 5a, 5b), thus the extracted components account for a substantial proportion of the variable's variance. This means that these variables are reflected well via the extracted components, and hence that the PCA analysis was reliable.

424 3.2 A quantitative perceptual model for estimating soil redistribution rates and the 425 effect of linear landscape elements

426 Assuming that each transect represents a 1 m wide strip, values of soil redistribution 427 rate obtained for each sampling point were used to calculate equivalent values of soil loss or deposition (kg year⁻¹) for individual slope segments, extending halfway to the 428 429 adjacent coring points from the sampling point in each direction, as reported by Collins 430 et al. (2001), Walling et al. (2003) and Estrany et al. (2010). However, this methodology 431 was modified to adapt it to the characteristics of the study transects, in order to take into 432 account changes in land use and the presence of linear landscape elements. This was 433 achieved by relating the segment length to vegetation cover and introducing the linear 434 landscape elements.

435 The resulting values for each segment were summed to provide a total erosion and total deposition, respectively, thus obtaining net soil loss for each transect (Mg ha⁻¹ year⁻¹) 436 437 and the sediment delivery ratio (%) (cf. Walling et al., 2003). For NT, the total erosion is estimated at 26 kg year⁻¹ and total deposition at 25.8 kg year⁻¹. The net soil loss of 0.2 438 kg year⁻¹ (0.01 Mg ha⁻¹ year⁻¹ and a sediment delivery ratio of 0.7 %) indicates that soil 439 440 redistribution processes have a limited effect on this transect. The presence of a stone 441 embankment between NT-4 and NT-5, and an unpaved trail between NT-6 and NT-7 is 442 likely to modify the runoff and sediment connectivity along the transect, except during 443 intense rainfall events. In contrast, for ST the total erosion is estimated to be 637 kg year⁻¹ and total deposition at 705 kg year⁻¹, representing a net soil accumulation of 68 kg 444 year⁻¹ (1.24 Mg ha⁻¹ year⁻¹ and a negative sediment delivery ratio), with this especially 445 446 concentrated along the bottom part of the ST.

447 Transect ST is characterized by the presence of a thick Muschelkalk outcrop at the448 midslope, between ST-13 and ST-15, which disrupts the runoff and sediment

449 connectivity along the transect. In addition, an unpaved trail located on the bottom slope 450 (between ST-24 and ST-25), a system of old terraces located in the upper part of the 451 transect (between ST-10 and ST-11) and several vegetation strips (Figure 5), also 452 modify the topography and change the runoff and sediment connectivity along the 453 transect.

454 Considering the natural elements and human modifications, mentioned above, transect 455 ST was divided into seven sections (Figure 5). During normal rainfall events the linear 456 landscape elements restrict the runoff and the downslope transfer of soil previously 457 eroded within each of the seven sections. However, during intense and erosive rainfall 458 events only the thick outcrop disrupts the soil redistribution processes along ST. In the 459 upper part of the transect, the vegetation cover on uncultivated areas is dense and, in 460 spite of the presence of the steepest slopes, the first three sections recorded low values of net soil loss (0.3, 0.3, 2.8 Mg ha⁻¹ year⁻¹, respectively, and sediment delivery ratios of 461 462 63, 100 and 100 %, respectively). In sections four and five higher values of net soil loss coincide with cultivated soils on steep slope (54.2 and 12.9 Mg ha⁻¹ year⁻¹, respectively, 463 464 and the corresponding sediment delivery ratios were 100 %). Section six is 465 characterized by low erosion rates on uncultivated areas and net soil deposition (6.5 Mg ha⁻¹ year⁻¹), which occurs in the cultivated fields above the trail. The last sections 466 467 correspond with cultivated flat areas below the trail, with higher net soil deposition $(1.91 \text{ Mg ha}^{-1} \text{ year}^{-1}).$ 468

This methodology provides information regarding erosion and deposition rates, as well
as the net soil loss from the transects, and how land use and linear landscape elements
modify the soil redistribution processes and sediment connectivity.

The patterns of ²¹⁰Pb_{ex} redistribution along both NT and ST demonstrate that in
Mediterranean environments cultivated land exerts an important control on soil loss

474 stressing the need to encourage the participation of the farmers in soil conservation 475 programs. Furthermore, these results suggest that deposition rates associated with 476 cultivated areas are affected by the presence of flat topography and soil conservation 477 practices, while deposition rates on uncultivated areas are linked to changes from 478 convex to concave slopes, the presence of transverse terraces and vegetation buffer 479 strips, which reduce runoff velocity. These results emphasize the potential of the new 480 green areas program proposed by CAP to control soil loss in agricultural ecosystems.

481 Sediment mobilised from the upslope areas may be deposited in the Estaña lake located 482 downslope of the investigated transects. The net soil deposition rates obtained for NT 483 and ST are influenced by the location of the soil sampling sites selected along 1 m wide strip. However, previous research using ¹³⁷Cs measurements (Gaspar et al., 2013) 484 showed high activity of ¹³⁷Cs in deeper layers at a sampling point situated on the margin 485 486 of the Estaña lake, adjacent to ST-27 at the bottom of the ST. This profile corresponds to a lake sediment deposit, as indicated by the presence of the 1963 ¹³⁷Cs peak at a 487 depth of 45 cm, which means an accumulation sediment of 113 Mg ha⁻¹ year⁻¹ at this 488 489 point.

490

491 **4 Conclusions**

This study has demonstrated the potential of 210 Pb_{ex} measurements to estimate soil erosion and deposition along the toposequences that are characteristic of hillslopes in mountain Mediterranean agroecosystems. For intricate transects, the sampling strategy should take into account changes in land use and the presence of linear elements in cultivated fields that might intensify tillage erosion at the head of the fields. For transects with homogeneous land use, a spacing of 50 m between sampling points is considered sufficient to provide meaningful estimates of soil redistribution rates. This 499 contribution describes similar soil redistribution patterns along the toposequences to 500 those established using 137 Cs, 210 Pb_{ex} and prediction models in previous research.

501 The spatial variability of soil redistribution rates along the toposequences was closely 502 controlled by land use that was in turn closely related to vegetation cover, topography, 503 soil type and slope gradient. Our results show that on steep cultivated slopes erosion 504 processes predominated, whereas uncultivated areas were characterized by lower soil 505 redistribution rates. On the flat areas at the bottom of both transects, sedimentation 506 processes dominated over erosion. The marked variations of SOC content along the 507 transect clearly reflect the variety of land use along the transects and their complex 508 physiography.

Land use and slope gradient exert important controls on the soil redistribution rates. For steep slopes on the upper part of the transects, the open Mediterranean forest and scrubland protect the soil surface from erosion, while the cultivated soils are more vulnerable to erosion and soil redistribution is more intense. Vegetation cover together with topography and tillage are key factors affecting the pattern of soil redistribution on the transects.

Assessing erosion and deposition rates for cultivated and uncultivated soils has proved useful for understanding the dynamics of soil redistribution in mountain agroecosystems. The application of a quantitative perceptual model has provided information to assess the effects of linear landscape elements along complex toposequences. This research has contributed information on the potential role of linear landscape elements and vegetation buffer strips in controlling sediment transfer along hillslopes within Mediterranean agroecosystems.

522

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637 Tables

638

Table 1. Basic statistics of 210 Pb_{ex} inventories (Bq m⁻²), and the main physicochemical soil properties for cultivated and uncultivated soils. Different letters indicate significant differences at the p-level < 0.05 between cultivated and uncultivated soils.

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		Cult	ivated n=9)		Uncultivated n=24				
-	Min.	Max.	Mean	SD	Min.	Max.	Mean		SD	
210 Pb _{ex} Bq m ⁻²	b.d.l.	7298.2	2975.9	a	2904.4	325.7	5729.4	1933.2	a	1042.4
SOC %	0.6	2.3	1.2	а	0.6	1.0	10.1	4.6	b	2.6
Stoniness %	5.0	55.9	27.0	a	16.6	20.6	70.4	45.3	b	12.6
Clay %	15.6	37.4	24.5	a	6.1	11.9	40.8	23.4	а	5.4
Silt %	35.7	77.6	63.5	a	13.4	41.9	78.6	63.8	а	8.6
Sand %	0.5	48.7	12.0	а	16.4	0.1	46.2	12.8	a	11.2

644 SD standard deviation

645 b.d.l. *below detection limit*

Table 2. Mean values of ²¹⁰Pb_{ex} inventories (Bq m⁻²) and the main physicochemical soil
properties for different soil types in cultivated and uncultivated soils.

		Cultivated				Uncultivate	d
		Calcisols	Regosols	Gypsisols	Leptosols	Calcisols	Regosols
		n=1	n=4	n=4	n=3	n=14	n=7
210 Pb _{ex} Bq m ⁻²	Mean	1654.6	1406.1	4876.0	1809.5	2056.2	1740.4
	SD	-	1837.0	3230.8	286.6	1208.5	942.2
SOC %	Mean	2.3	1.1	0.8	8.0	3.9	4.4
	SD	-	0.5	0.2	1.8	1.8	3.2
Stoniness %	Mean	48.8	33.1	15.2	53.9	41.7	49.0
	SD	-	16.7	7.7	2.9	12.5	13.7
Clay %	Mean	21.3	21.3	28.5	23.9	24.2	21.8
	SD	-	5.1	6.0	2.7	6.6	3.4
Silt %	Mean	49.9	64.4	66.1	67.1	60.3	69.2
	SD	-	19.5	5.1	2.5	7.7	9.2
Sand %	Mean	28.8	14.3	5.4	9.1	15.5	9.0
	SD	-	22.9	5.3	5.0	12.0	10.7

651 SD standard deviation

Table 3. Summary statistics of soil erosion and deposition rates (Mg ha⁻¹ year⁻¹) for
sampling sites on cultivated and uncultivated soils.

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Mg ha ⁻¹ year ⁻¹		Median	Mean	SD	SE	Min.	Max.	CV %
Erosion								
Cultivated	5	12.9	32.1	35.4	15.8	4.2	83.7	110.3
Uncultivated	15	0.5	0.9	0.8	0.2	0.1	2.4	89.1
Deposition								
Cultivated	4	56.5	54.1	20.6	10.3	28.6	74.75	38.1
Uncultivated	9	0.5	1.1	1.8	0.6	0.1	5.6	162.2

656 SD standard deviation

657 SE standard error

Table 4. Multiple range test for soil erosion and deposition rates (Mg ha⁻¹ year⁻¹) associated with different edaphic and physiographic characteristics. Different letters indicate significant differences at the p-level < 0.05 between different land uses, soil types and slope gradients, respectively.

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	Eros	sion rate	Mg	ha ⁻¹ year ⁻¹	Deposition rate Mg ha ⁻¹ year ⁻¹			
	n	Mean		SD	n	Mean		SD
Land use								
Uncultivated	15	0.9	a	0.8	9	1.1	а	1.8
Cultivated	5	32.1	b	35.4	4	54.1	b	20.6
Soil type								
Leptosols	2	0.5	a	0.3	1	0.1	a	-
Calcisols	10	1.2	a	1.6	5	1.5	a	2.3
Regosols	7	10.9	a	19.6	4	7.7	a	13.9
Gypsisols	1	83.7	b		3	62.6	b	14.3
Slope %								
0-12	9	10.5	a	275	6	37.2	b	30.7
12-24	7	10.6	a	19.8	5	0.3	а	0.2
> 24	4	1.1	a	0.9	2	0.8	ab	0.8

664 SD standard deviation

Table 5.a. Varimax rotated principal component loading (PCi) for the three first
components (erosion rates). Loading factors higher than 0.5 (absolute value) are shown
in bold.

				Estimated
	PC1	PC2	PC3	communality
Use	0.31186	0.83757	0.02315	0.79931
Stoniness %	0.18808	-0.63599	0.18469	0.47397
Erosion rates Mg ha ⁻¹ year ⁻¹	-0.05773	0.86661	-0.07046	0.75931
Clay %	-0.75604	0.07373	0.02519	0.57767
Silt %	-0.83205	-0.03449	-0.16506	0.72074
Sand %	0.97829	-0.00531	0.11993	0.97147
SOC %	0.11328	-0.66614	-0.58696	0.80110
Slope %	0.20854	-0.20802	0.83489	0.78381

Table 5.b. Varimax rotated principal component loading (PCi) for the two first
components (deposition rates). Loading factors higher than 0.5 (absolute value) are
shown in bold.

			Estimated
	PC1	PC2	communality
Use	-0.91573	0.28580	0.92023
Stoniness %	0.81778	-0.24505	0.72882
Deposition rates Mg ha ⁻¹ year ⁻¹	-0.92483	0.18565	0.88977
Clay %	-0.37995	0.59488	0.49824
Silt %	0.06915	0.91820	0.84788
Sand %	0.12374	-0.97653	0.96893
SOC %	0.61104	-0.61082	0.74646
Slope %	0.77786	0.16928	0.63373

678 Figures

Figure 1. The study area located in the northern border of central part of the Ebro basin
(NE Spain) and the 34 sampling sites situated along southern (ST) and northern (NT)
transects.





Figure 3. Estimates of soil redistribution rates based on the 210 Pb_{ex} inventory measurements for the individual sampling points along northern (NT) and southern (ST) transect. Black numbers indicate cultivated soil profiles and grey numbers indicate uncultivated soil profiles.



Figure 4. PCA biplot: dispersion diagram and principal components loadings, PC
loading 1 vs. PC loading 2 of cultivated and uncultivated soil samples after PCA
Varimax rotated for a) erosion rates and b) deposition rates.



700 Figure 5. Estimates of soil redistribution rates for individual slope segments and net soil



