Fate of $^{210}\text{Pb}_{\text{ex}}$ fallout in soil under forest and scrub of the central Spanish Pre-Pyrenees

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Running title: The $^{210}\text{Pb}_{\text{ex}}$ profiles in forest and scrub soil

Highlights:

- Characterizing the vertical distribution of $^{210}\text{Pb}_{\text{ex}}$ requires high resolution sampling (2 cm)
- In uncultivated soil of the Pre-Pyrenees the fallout, $^{210}\text{Pb}_{\text{ex}}$, is retained in the uppermost 14 cm
- The distribution of $^{210}\text{Pb}_{\text{ex}}$ matches that of the organic matter down the profile
- Vegetation affects the surface activity of $^{210}\text{Pb}_{\text{ex}}$ in the soil, being greater under forest than under scrub.

Summary

The characteristics and distribution of unsupported radioactive lead-210 ($^{210}\text{Pb}_{\text{ex}}$) in soil and the relations between the radionuclide and soil properties determine its fate within the environment. We have explored the distribution of $^{210}\text{Pb}_{\text{ex}}$ in stony soil profiles near the edge of the Ebro basin in northern Spain, the role played by vegetation in that distribution, and the relation between the radionuclide and organic carbon. We describe in detail the profiles of $^{210}\text{Pb}_{\text{ex}}$ at 23 sites; 10 under forest and 13 under scrub, which were sampled at 2-cm intervals to a maximum depth of 14 cm. The theoretical distribution of $^{210}\text{Pb}_{\text{ex}}$ follows an exponential decline with depth in undisturbed soil, assuming minimal surface slope and no evidence of erosion or deposition processes. Comparable distribution patterns of $^{210}\text{Pb}_{\text{ex}}$ and organic C became evident from the analyses. There were significant correlations between the activity of $^{210}\text{Pb}_{\text{ex}}$ and organic C under both forest and scrub, but the strongest were in the surface layers of soil under forest. More than 80% of the total activity of $^{210}\text{Pb}_{\text{ex}}$ was adsorbed in the upper 6 cm, with an exponential decline with increasing depth. The decline was modelled with exponential functions fitted by non-linear least-squares regression to predict the depth distributions of $^{210}\text{Pb}_{\text{ex}}$ and organic C in forest and scrub soils separately. The results confirm the viability of the use of $^{210}\text{Pb}_{\text{ex}}$ in stony soil as an indicator of soil redistribution and show the significant effect of vegetative cover. These results provide useful information about sampling design for future research if the radionuclide $^{210}\text{Pb}_{\text{ex}}$ is used for assessing soil redistribution in similar Mediterranean environments.

Introduction

The radionuclide lead-210 (^{210}Pb) is among the most important environmental radionuclides. It is a member of the uranium-238 (^{238}U) decay series and is an important component of the natural background radiation. It occurs naturally and in varying concentrations in soil, sediments and natural water. Lead-210 (with a half-life of 22.3 years) derives from the decay of radon-222 (^{222}Rn) and the daughter of radium-226 (^{226}Ra), which occur naturally in soil and rocks. The ^{210}Pb in soil generated *in situ* by the decay of ^{226}Ra is termed ‘supported’ ^{210}Pb ($^{210}\text{Pb}_s$), which is in equilibrium with ^{226}Ra . Upward diffusion of a small quantity of ^{222}Rn from the soil introduces ^{210}Pb into the atmosphere. Subsequently this ^{210}Pb falls back to the surface of the soil through wet and dry deposition from the atmosphere (Figure 1). This deposited ^{210}Pb is not in equilibrium with its parent ^{226}Ra , and is commonly termed ‘unsupported’ or ‘excess’ ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$). Thereafter $^{210}\text{Pb}_{\text{ex}}$ is rapidly and strongly sorbed by the soil (He & Walling, 1996), although some moves downwards in the profile. Considerable variation in the global pattern of $^{210}\text{Pb}_{\text{ex}}$ fallout has been documented. Appleby & Oldfield (1992) pointed out that values are greater in eastern parts of continents because of the predominant west-east movement of air masses, and are less over the oceans (Walling & He, 1999).

The vertical distribution of $^{210}\text{Pb}_{\text{ex}}$ is controlled primarily by re-distribution processes and is affected by surface runoff, mixing by bioturbation and vertical migration. In stable, undisturbed soil the activity of $^{210}\text{Pb}_{\text{ex}}$ decreases approximately exponentially with depth (Figure 1). The adsorption of ^{210}Pb in soil is affected by various properties, such as clay and organic matter (Fujiiyoshi & Sawamura, 2004), and soil type (Özden *et al.*, 2013). Previous research on soil in the Spanish Pyrenees indicated that the distribution of ^{210}Pb is related to carbonates and to iron (Fe) and manganese (Mn) oxides, whereas variation in ^{238}U seems to be

more related to processes such as leaching and the transport and accumulation of soil particles (Navas *et al.*, 2005, 2011). In addition, the $^{210}\text{Pb}_{\text{ex}}$ profiles in soil under forest are probably affected by the canopy (Kato *et al.*, 2010; Teramage *et al.*, 2015), which intercepts $^{210}\text{Pb}_{\text{ex}}$ fallout, and depends to some degree on the stage of vegetation growth and geometry of the foliage.

Recently, Mabit *et al.* (2014) reviewed the environmental behaviour of ^{210}Pb and emphasized the importance of soil physicochemical characteristics such as texture, pH, organic matter and cation exchange capacity (CEC) for the bioavailability of Pb in soil. These physicochemical properties play an important role in the amount of ^{210}Pb in the environment (Narayana & Rajashekara, 2010).

There have been a few attempts to explore the vertical distribution of unsupported ^{210}Pb in soil profiles in Mediterranean environments (Ugur *et al.*, 2004; Benmansour *et al.*, 2011; Porto & Walling, 2012). Our earlier research in the Spanish Pre-Pyrenees combined the use of both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ to assess rates of soil redistribution (Gaspar *et al.*, 2013a,b) and showed the potential of $^{210}\text{Pb}_{\text{ex}}$ as a sediment radiotracer (Gaspar *et al.*, 2013a). Furthermore, we stressed the need to define better a suitable depth of sampling and sampling intervals in heterogeneous environments with an intricate mosaic of land uses as in the Pre-Pyrenean agroecosystems (Gaspar *et al.*, 2013b). This encouraged our further research on the depth distribution of the radionuclide and the main relations with soil properties under natural vegetation in stony mountain soil.

We report here our further exploration of the distribution and behaviour of $^{210}\text{Pb}_{\text{ex}}$ in stony soil of the central Spanish Pre-Pyrenees, and how the variation in $^{210}\text{Pb}_{\text{ex}}$ is affected by vegetation cover, soil depth and the contents of soil organic carbon (SOC) and stone.

Our main objective was to discover whether there were differences in $^{210}\text{Pb}_{\text{ex}}$, both its activity and inventory, in soil between forest and scrub and the relation of $^{210}\text{Pb}_{\text{ex}}$ to depth in the soil. To this end we (i) describe the vertical distributions of $^{210}\text{Pb}_{\text{ex}}$ and organic C, (ii) use exponential regression models for assessing the effect of vegetation cover and soil depth on $^{210}\text{Pb}_{\text{ex}}$ and organic C and (iii) explore the relations between $^{210}\text{Pb}_{\text{ex}}$ with soil organic C and stone content.

Sampling and measurements

Study area

The study was carried out on vegetated hillslopes that are representative of Mediterranean mountains in the central Spanish Pre-Pyrenees ($42^{\circ} 1' 60''\text{N}$, $0^{\circ} 31' 8''\text{E}$) (Figure 2). The main soil types are Leptosols, Calcisols and Regosols (see Gaspar *et al.*, 2014). The vegetation comprises Mediterranean forest and scrub, which correspond with the Mediterranean and sub-Mediterranean bioclimatic regimes. The forest is dominated by evergreen oak (*Quercus rotundifolia* Lam.) and drought-resistant deciduous oaks (*Quercus faginea* Lam. and *Quercus cerrioides* Willk & Costa), and there is a large percentage of litter cover and leaves on the soil surface. Scrub is dominated by some *Buxus sempervirens* L., *Juniperus oxycedrus* L. and sub-Mediterranean plants, which cover less of the ground. The climate is continental Mediterranean with an average annual rainfall of 595 mm distributed in two rainy seasons, spring and autumn, and a mean annual temperature of 12.2°C . An accurate characterization of soil properties at the sampling sites was made for the soil samples taken from forest and scrub areas.

Measurement of soil properties and the data

Twenty-three soil profiles were chosen at random locations for sampling; they included 10 in forest and 13 under scrub. The profiles were on sites with gradients that ranged from moderate (0–12%) to medium (12–24%) and steep (>24%). The soil types sampled included three Leptosols, four Calcisols and three Regosols in the forest, and nine Calcisols and four Regosols in scrub (Table 1).

In an earlier investigation in the region (Gaspar *et al.*, 2013b) we sliced vertical cores of soil into 5-cm sections in which to measure $^{210}\text{Pb}_{\text{ex}}$. We found that the uppermost 14 cm of uncultivated soil contained 80% of the $^{210}\text{Pb}_{\text{ex}}$ activity, and that we had too few measurements to describe the distribution. Therefore, in our new investigation we sampled at 2-cm intervals to a maximum depth of 14 cm. We thought that this would enable us to obtain more detailed profiles of $^{210}\text{Pb}_{\text{ex}}$. The soil samples were taken with a 10 cm $\times$ 10 cm steel box corer (Figure 2) designed by Navas *et al.* (2008), which can sample smaller depth increments of soil. Although we could sample in most of the profiles to 14 cm, for some Leptosols the maximum sampling depth was ≤ 10 cm (P2) and ≤ 12 cm (P3) and in some Calcisols it did not extend below 12 cm (P14 and P23) because of underlying rock (Table 1). We took 156 soil samples in total (67 samples from forest and 89 from scrub profiles). These were air-dried, ground and sieved to pass 2 mm.

Measurements of $^{210}\text{Pb}_{\text{ex}}$ were made by gamma-ray spectrometry, with the high resolution low energy coaxial HPGe detector model XtRa GX3019 (Canberra, Meriden, CT, USA) coupled to an amplifier. The detector had a relative efficiency of 50% and a resolution of 1.9 keV (shielded to reduce background radiation) (see Navas *et al.*, 2013). After equilibrium between ^{226}Ra and its daughter ^{214}Pb was reached, the total ^{210}Pb activity was measured at 46.5 keV. We calculated unsupported $^{210}\text{Pb}_{\text{ex}}$ activity by subtracting ^{226}Ra from total ^{210}Pb . The

activity of ^{226}Ra was obtained by measurement of the activity of ^{214}Pb , a short-lived daughter of ^{226}Ra at 351.9 keV. Count times were typically of about 86 400 s, with an analytical precision of about 10–15% at the 95% level of confidence. The activity of $^{210}\text{Pb}_{\text{ex}}$ is expressed as concentration (Bq kg^{-1}) and the inventory (Bq m^{-2}) is the activity per unit area (1 m^2) calculated from the dry mass bulk density (g cm^{-3}) of the fine fraction ($< 2 \text{ mm}$) for each layer. The total inventory of the profile would equal the sum of the 2-cm slices.

The contents of soil organic carbon (SOC) and soil inorganic carbon (TIC) were measured by dry combustion with a LECO RC-612 multiphase carbon analyser (LECO, St. Joseph, USA). Stone content was defined as the percentage of fraction greater than 2 mm, and density of the fine fraction ($< 2\text{mm}$) was determined from the known total volume of the sample and dry weight of the fine fraction, on the assumption that the density of stones was 2.63 g cm^{-3} . The particle-size distribution was determined by laser diffraction (Beckman Coulter LS 13 320, Miami, FL, USA) after chemical elimination of the organic matter and chemical disaggregation. The pH (in a 1:2.5 soil:water suspension) and electrical conductivity (EC) (in a 1:5 soil:water suspension) were measured by standard techniques.

We analysed the data statistically using Statgraphics for Windows (Statgraphics Centurion XVII, 2016) and GenStat (Payne, 2016).

Statistical analysis and results

Summary statistics and analysis of variance

The soil samples ($n = 156$) are very stony in general and their texture is predominantly silty loam (Table 1). The silt fraction is predominant and ranges from 14 to 80%, and the clay content

of 88% of samples shows moderate variability, between 15 and 30 %. The sand content is less than 10% for 80% of the samples, although it reaches 84% in some. The SOC varies considerably from 0.61 to 29%. The density of the <2mm fraction has a minimum value of 0.20 g cm⁻³ in organic soil and reaches a maximum of 2.31 g cm⁻³. The soils are alkaline (pH: 6.89–8.17) with little evidence of salinity (electrical conductivity, EC: 0.185–0.718 dSm⁻¹) and calcareous with ranges of total inorganic carbon from 0.40 to 12.8 %. The ²¹⁰Pb_{ex} activity varies considerably; it reaches a maximum of 195.6 Bq kg⁻¹ with a median of 23.3 Bq kg⁻¹ and a minimum less than detection limit. The inventory of ²¹⁰Pb_{ex} ranges from zero to 3026.0 Bq m⁻².

The forest soil has the largest ²¹⁰Pb_{ex} activity and SOC. The maximum values of SOC are in the surface layers of forest soil, whereas for scrub soil the largest SOC content is about half that of the forest values (11 %). The stone content is larger for the forest soil, which has a smaller density in the fine fraction (<2 mm), with a maximum that does not exceed 1.59 g cm⁻³. Conversely, soil under scrub generally recorded a slightly lower ²¹⁰Pb_{ex} activity, but the ²¹⁰Pb_{ex} inventories deviated more than the reference inventory for the area (2019 ± 215.8 Bq m⁻², Gaspar *et al.*, 2013a). The fine fraction (<2 mm) of the scrub soil also has the largest density. The pH is similar for forest and scrub soil, but we consider that leaching might be more intense in Leptosols (forest profiles P1, P2, P3) because the maximum pH does not exceeded 7.85. This is less than the pH of Calcisols and Regosols, which are the soil types that predominate under scrub. Figures S1 and S2 (Supporting Information) provide detailed information for all soil properties analysed.

The data for ²¹⁰Pb_{ex}, both their activity and inventory, the contents of organic C (SOC) and stone and the bulk densities are displayed layer by layer in Figure 3 as box-and-whisker diagrams. It is apparent that the largest concentrations of ²¹⁰Pb_{ex} and organic C are in the

uppermost 2 cm and the concentrations decrease monotonically down the profiles. We return to this matter below. Apart from the clear decrease in stone content in the three first depth intervals, there are no evident trends in either the stone content or bulk density.

From the data on the 2-cm slices we estimated the amounts of $^{210}\text{Pb}_{\text{ex}}$, organic C and stone in the soil down to 14 cm under forest and scrub vegetation, and compared them by an analysis of variance. For this we calculated the amounts of C and stone per square metre, knowing the dry mass fine fraction per square metre of the individual 2-cm slices as for the $^{210}\text{Pb}_{\text{ex}}$ inventory. Figure 4 summarizes the results graphically; the difference in carbon is substantial with a mean of 5.09 kg m² under forest compared with a mean of 3.40 kg m² under scrub. Perhaps surprisingly, there is little difference in the $^{210}\text{Pb}_{\text{ex}}$ inventories for the forest and scrub sites. The mean values for stone content are almost identical. The results of the analyses of variance are summarized in Table 2.

Modelling the vertical distribution of $^{210}\text{Pb}_{\text{ex}}$ and organic C

Figure 3 suggests that both $^{210}\text{Pb}_{\text{ex}}$ activity and the concentration of organic C decrease exponentially down the profiles in accord with what other investigators have observed. Nevertheless, the distributions appear to differ substantially from one another in magnitude. To pursue aim (ii), i.e. the comparison of these distributions, we fitted exponential models to them, taking into account that we had repeated measurements on the same profiles.

The basic equation for the distribution is

$$y = \alpha + \beta \times \omega^x, \quad (1)$$

where y is the response variable, here the $^{210}\text{Pb}_{\text{ex}}$ activity or the concentration of organic C, and x is the depth. The quantities α , β and ω are fitting parameters: α is the asymptote to which y

approaches as x , the depth, increases, $\alpha + \beta$ is the maximum of y at $x = 0$, the surface of the soil, and ω , which must be less than 1, describes the curvature.

To compare the equations for forest and scrub we elaborate, defining two equations:

$$^{210}\text{Pb}_{\text{ex, forest}} = A_{\text{forest}} + B_{\text{forest}} \times W_{\text{forest}}^x, \quad (2)$$

and

$$^{210}\text{Pb}_{\text{ex, scrub}} = A_{\text{scrub}} + B_{\text{scrub}} \times W_{\text{scrub}}^x, \quad (3)$$

where $^{210}\text{Pb}_{\text{ex}}$ is in Bq kg^{-1} and x is in cm and in which the parameters A , B and W represent α , β and ω , respectively, and are specific to forest and scrub separately. The equations for organic C are analogous.

Our task became that of fitting simultaneously the equations for the two kinds of vegetation as factors in terms of the three parameters, and of testing the parameter estimates against the null hypothesis that they were not different. We discovered that we needed to transform the measured values to square roots to normalize the residuals and to make the residuals independent of one another. All the analyses, therefore, were done on square roots. The latter and the values of $^{210}\text{Pb}_{\text{ex}}$ and organic C predicted by the fitted equations were back-transformed for display in Figure 5. To take into account the repetition of measurements on the same profiles we incorporated a first-order autoregressive dependence within each of the profiles.

We proceeded in steps, first fitting Equation (1) to all the data regardless of the vegetation, and then adding separate constants A_{forest} and A_{scrub} , then separate B_{forest} and B_{scrub} , and finally separate W_{forest} and W_{scrub} . Tables S1 and S2 (Supporting Information) lists the results of the analyses of variance and parameter estimates at every step in the analysis. Tables 3 and 4 summarize the final analysis for $^{210}\text{Pb}_{\text{ex}}$ and organic C respectively, and Figure 5 shows the

functions fitted to both the $^{210}\text{Pb}_{\text{ex}}$ activities and concentrations of organic C under the two kinds of vegetation.

As expected, depth is very significant ($P < 0.001$) for both $^{210}\text{Pb}_{\text{ex}}$ and organic C under both forest and scrub. So too are the interactions, depth $\times$ vegetation, although somewhat less so for organic C than $^{210}\text{Pb}_{\text{ex}}$. The maxima too of the functions (A + B) differ significantly.

Vegetation on its own was very significant in relation to the difference in organic C between forest and scrub (Table 4), but the difference in $^{210}\text{Pb}_{\text{ex}}$ was not (Table 3). The non-linear shape parameters W_{forest} and W_{scrub} were similar for both $^{210}\text{Pb}_{\text{ex}}$ and organic C, and not significantly different.

Relations between $^{210}\text{Pb}_{\text{ex}}$ and soil properties

We discovered that most of the $^{210}\text{Pb}_{\text{ex}}$ is retained in the uppermost 2 cm of the profiles, which is also richest in organic C. The Pearson correlation coefficient, computed on the square roots of the $^{210}\text{Pb}_{\text{ex}}$ activity and organic C, is 0.885 for the forest profiles and 0.526 for the scrub (Table 5).

The $^{210}\text{Pb}_{\text{ex}}$ activity is significantly and positively correlated also with stone content for forest and scrub soil (Table 5), whereas the correlation between $^{210}\text{Pb}_{\text{ex}}$ activity and density of the fine fraction (<2 mm) is significant and negative at the 0–2-cm depth interval, especially for the forest soil.

We calculated also the correlation coefficients between the $^{210}\text{Pb}_{\text{ex}}$ inventory (in Bq m^2) and the mass of organic C and stone contents to the same maximum depth (in kg m^2), but there were no significant correlations.

Discussion

In the sample profiles the uppermost 6 cm contains more than 80% of $^{210}\text{Pb}_{\text{ex}}$, and there is none below 12 cm. These results accord with those from previous studies in which all $^{210}\text{Pb}_{\text{ex}}$ activity was found to occur in the upper 16 cm (Walling *et al.*, 2003), 18 cm (Benmansour *et al.*, 2011) or 20 cm (Zheng *et al.*, 2007). Odzen *et al.* (2013) found that $^{210}\text{Pb}_{\text{ex}}$ can move to a maximum depth of about 20 cm in undisturbed soil. The continuous inputs of atmospheric fallout of unsupported ^{210}Pb to the soil surface have maintained a layer of maximum activity at the surface and accentuate the importance of the near-surface horizon in accounting for most of the $^{210}\text{Pb}_{\text{ex}}$. The pattern we observed almost certainly arises because of strong adsorption of the $^{210}\text{Pb}_{\text{ex}}$ by the surface soil, leaving little to be redistributed within the profile (Zhang *et al.*, 2006). This behaviour is similar to that reported by Walling & Quine (1995) for ^{137}Cs .

Infiltrating rain can transport the radionuclide down the soil profile (He & Walling, 1997) and lead to an exponential decrease in it with increasing depth. We (Gaspar & Navas, 2013) observed a similar behaviour with the artificial fallout of ^{137}Cs , although maximum ^{137}Cs activity occurred at 6 cm in uncultivated soil. Wakiyama *et al.* (2010) also obtained similar results; they observed that the maximum concentrations of $^{210}\text{Pb}_{\text{ex}}$ occurred at 1.5–2 cm, whereas that of ^{137}Cs content was greatest at 3.5–4.5 cm below the surface.

The larger concentrations of $^{210}\text{Pb}_{\text{ex}}$ in profiles with large stone contents are because the amount of adsorbed $^{210}\text{Pb}_{\text{ex}}$ per unit mass of soil surface is greater when there is less soil available in relation to the effective volume of the fine fraction that can retain fallout radionuclides (Soto & Navas, 2004).

In general, the decrease in content of $^{210}\text{Pb}_{\text{ex}}$ with depth in the forest and scrub soil reflects strong adsorption of the radionuclide by the surface soil during infiltration. In our karstic study area, however, the effect of pH on leaching of the nuclide cannot be totally excluded, which

accords with findings by Evans *et al.* (1997) for the glaciers from Conway Granite in Carroll County, New Hampshire (USA). In our shallow profiles pH changes little with depth (apart from profiles P7, P8, P17 and P23), but in the deeper profiles pH was higher in the deepest soil layers (see also Gaspar & Navas, 2013), probably because of the leaching of carbonates (Navas *et al.*, 2005).

The behaviour of $^{210}\text{Pb}_{\text{ex}}$ and its distribution with depth depend on pedogenic processes that include losses by leaching, which could be explained by karstic processes that partially control evolution of the landscape on which the Leptosols have developed (Navas *et al.*, 2013). Smith & Blake (2014) also identified a vertical control on soil properties under a uniform geological substrate in which the content of $^{210}\text{Pb}_{\text{ex}}$ is greater at the soil surface. In addition, losses and gains by plant uptake and subsequent decay of plant materials, especially in the A horizon, should be also considered as Evans *et al.* (1997) point out.

The large $^{210}\text{Pb}_{\text{ex}}$ activities and inventories at the soil surface suggest strong fixation by the fine components of the soil. Although fallout radionuclides are adsorbed on to clay surfaces or fixed within the lattice structure, we know that organic matter also has a greater effect (Rigol *et al.*, 2002). The fact that the exponential function fits the vertical distributions of both $^{210}\text{Pb}_{\text{ex}}$ activity and organic C so well suggests that organic matter is the major adsorbant of the $^{210}\text{Pb}_{\text{ex}}$ in the profiles we sampled. This inference accords with the results of our earlier investigation (Gaspar *et al.*, 2013b) in soil profiles with uniform distributions of clay. Fujiyoshi *et al.* (2011) also found that organic matter also plays a more important role than that of clay in determining the vertical distribution of $^{210}\text{Pb}_{\text{ex}}$, as did Sipos *et al.* (2005) and Degryse *et al.* (2009), especially in acidic soil. The greater affinity of $^{210}\text{Pb}_{\text{ex}}$ than ^{137}Cs for soil organic matter (Wallbrink *et al.*, 1998) and its accumulation in the litter layer associated with humic substances might retard the downward migration of $^{210}\text{Pb}_{\text{ex}}$ in comparison with ^{137}Cs (Dorr & Munnich, 1991).

The models for forest and scrub profiles illustrate the differences in the vertical distributions of $^{210}\text{Pb}_{\text{ex}}$ under the two kinds of vegetation, especially in the upper few centimetres. The strong similarity in distributions of organic C with those of $^{210}\text{Pb}_{\text{ex}}$ suggest the strong control of organic matter on the retention of $^{210}\text{Pb}_{\text{ex}}$. Models of organic C and $^{210}\text{Pb}_{\text{ex}}$ with depth support the evidence that these two properties decrease similarly down the profile and are strongly related, especially in forest soil.

The depletion of $^{210}\text{Pb}_{\text{ex}}$ in scrub profiles, indicated by the greater deviation of $^{210}\text{Pb}_{\text{ex}}$ from the reference inventory than in forest profiles (see Figure S1, Supporting Information), suggests that scrub profiles are more affected by soil redistribution processes. The number of soil profiles that show depletion of $^{210}\text{Pb}_{\text{ex}}$ and SOC under scrub are probably related to a smaller percentage vegetation cover in certain places. This results in less protection of the soil surface and there is probably more soil erosion, which would reduce both radionuclide content and SOC in the surface layers. By comparison, dense mature forest with greater cover would reduce the splash and detachment of soil particles and so limit $^{210}\text{Pb}_{\text{ex}}$ depletion. In other nearby temperate areas the forest soil was found to be more resistant to erosion than scrub soil (Navas *et al.*, 2008), in spite of the greater slope (Gaspar *et al.*, 2013a). Stable profiles were predominant in the scrub soil, which comprised > 50 % of the profiles analysed in this study. Therefore, as in other Mediterranean environments, lower growing shrubs and grasses might also be effective in protecting soil from erosion because they provide enough ground surface cover as Quine *et al.* (1994) also found in the central Ebro valley.

Conclusions

Our results show that the content of $^{210}\text{Pb}_{\text{ex}}$ below 14 cm was negligible and that 80% of the content of $^{210}\text{Pb}_{\text{ex}}$ occurs in the upper 6 cm of the soil profile. We recommend sampling at 2-cm depth intervals to improve the accuracy of $^{210}\text{Pb}_{\text{ex}}$ measurements in similar environments to that of this study.

The vertical distribution of $^{210}\text{Pb}_{\text{ex}}$ seems to be most affected by soil organic matter. Both the $^{210}\text{Pb}_{\text{ex}}$ and organic C appear to decline exponentially with increasing depth in the soil. In both the forest and scrub soil exponential functions fitted the observed distributions well. The parameters of the exponential functions fitted to the soil under forest, however, differed from those for the soil under scrub, and the differences show that vegetation cover plays an important role in the fate of $^{210}\text{Pb}_{\text{ex}}$ (Bq kg^{-1}).

We improved our understanding of how the radionuclide $^{210}\text{Pb}_{\text{ex}}$ is distributed in mountain soils in northern Spain, which is essential for interpreting the information that $^{210}\text{Pb}_{\text{ex}}$ provides as a tracer of soil erosion and deposition, as well as a fingerprinting of sources of sediment.

Supporting Information

Figure S1. Detailed information for all soil properties analysed in Figure 3 for each section of the profile, including the total inventory for each profile to compare with Reference inventory of the study site 2019 Bq m^{-2} .

Figure S2. Box-and-whisker plots of total inorganic carbon (TIC), pH, electric conductivity (EC) and clay, silt and sand contents for depth increments under (a) forest and (b) scrub.

Table S1. Detailed analysis of variance of $^{210}\text{Pb}_{\text{ex}}$ activity (Bq kg^{-1}) transformed to square roots for all steps in the analysis. Step 4 corresponds with Table 3.

Table S2. Detailed analysis of variance of organic C (%) transformed to square roots: for all steps in the analysis. Step 4 corresponds with Table 4.

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Figure Captions

Figure 1. (a) A compilation of ^{210}Pb knowledge. Decay scheme of ^{226}Ra and its daughters, the use of $^{210}\text{Pb}_{\text{ex}}$ measurements to estimate soil erosion and deposition rates and for identifying source and sink of sediments and (b) different examples of the theoretical vertical distribution of $^{210}\text{Pb}_{\text{ex}}$ in soil. This figure is a re-edition based on Walling *et al* (2003), completed by $^{210}\text{Pb}_{\text{ex}}$ research to date.

Figure 2. (a) Location of the study site, (b) detailed information of the high-resolution soil profile sampler and (c) characteristics of the forest and scrub areas in stony Mediterranean mountain soil (Northern Spain).

Figure 3. Box-and-whisker plots of $^{210}\text{Pb}_{\text{ex}}$, organic C and other soil properties for depth increments under (a) forest and (b) scrub.

Figure 4. Box-and-whisker plots of the total inventories of $^{210}\text{Pb}_{\text{ex}}$, organic C and stone content in the soil of the two vegetation types.

Figure 5. Vertical distributions of (a) $^{210}\text{Pb}_{\text{ex}}$ activity and (b) organic C under forest and scrub. The measurements are plotted as points, and the fitted functions, the parameters of which are listed in Tables 3 and 4, are shown by the curves.

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(a) Forest

(b) Scrub

Forest soil profiles n = 10
Scrub soil profiles n = 13

Q1 25 percentile
median
Whisker 10 percentile
Q3 75 percentile
Whisker 90 percentile
outlier

Figure 4. Box-and-whisker plots of the total inventories of $^{210}\text{Pb}_{\text{ex}}$, organic C and stone content in the soil of the two vegetation types.

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Supplementary Information 1. Vertical distributions of (a) $^{210}\text{Pb}_{\text{ex}}$ activity and (b) organic C under forest and

Supplementary Information 2.

(a) Forest

(b) Scrub

Table 1. Characterization of the 23 soil profiles sampled under forest and scrub Mediterranean mountain soil. Profiles were sampled at 2-cm depth increments

Forest: 10 profiles and 67 soil samples				
Profiles	Depth /cm	<i>n</i>	Soil Type	Texture
P1	14	7	Leptosol	Silty loam
P2	10	5	Leptosol	Silty loam
P3	12	6	Leptosol	Silty loam
P4	14	7	Calcisol	Silty clay loam
P5	14	7	Calcisol	Medium loam
P6	14	7	Calcisol	Silty loam
P7	14	7	Calcisol	Silty loam
P8	14	7	Regosol	Silty loam
P9	14	7	Regosol	Silty loam
P10	14	7	Regosol	Silty loam
Scrub: 13 profiles and 89 soil samples				
Profiles	Depth /cm	<i>n</i>	Soil Type	Texture
P11	14	7	Calcisol	Sandy loam
P12	14	7	Calcisol	Silty loam
P13	14	7	Calcisol	Silty loam
P14	12	6	Calcisol	Silty clay loam
P15	14	7	Calcisol	Silty clay loam
P16	14	7	Regosol	Medium loam
P17	14	7	Regosol	Silty loam
P18	14	7	Regosol	Silty loam
P19	14	7	Regosol	Silty loam
P20	14	7	Calcisol	Silty loam
P21	14	7	Calcisol	Silty clay loam
P22	14	7	Calcisol	Silty loam
P23	12	6	Calcisol	Silty clay loam

Depth, final depth of each profile; *n*, total number of soil samples for each profile

Table 2. Analysis of variance of $^{210}\text{Pb}_{\text{ex}}$, organic carbon and stone inventories

Source	Degrees of freedom	$^{210}\text{Pb}_{\text{ex}} / \text{Bq m}^{-2}$			SOC / kg m^{-2}			Stone / kg m^{-2}		
		Mean square	<i>F</i>	<i>P</i>	Mean square	<i>F</i>	<i>P</i>	Mean square	<i>F</i>	<i>P</i>
Vegetation	1	100 465	0.21	0.648	16.14	4.85	0.039	10.81	0.01	0.911
Residual	21	469 319			3.332			838.7		
Total	22	452 553								

Means and standard errors

	Vegetation	<i>n</i>	Mean	Standard error
$^{210}\text{Pb}_{\text{ex}} / \text{Bq m}^{-2}$	Forest	10	1693	±216
	Scrub	13	1826	±190
SOC / Kg m^{-2}	Forest	10	5.09	±0.57
	Scrub	13	3.40	±0.51
Stone / Kg m^{-2}	Forest	10	91.96	±9.16
	Scrub	13	93.35	±8.03

Table 3. Analysis of variance of $^{210}\text{Pb}_{\text{ex}}$ activity transformed to square roots

Source	Degrees of freedom	Mean square	<i>F</i>	<i>P</i>
Depth	2	481.2	154.7	<0.001
Vegetation	1	7.85	2.52	0.114
Depth $\times$ Vegetation	1	24.93	8.02	0.005
Separate non-linear <i>W</i>	1	2.36	0.76	0.385
Residual	150	3.11		
Total	155	9.45		

Parameter estimates and their standard errors

Parameter	Estimate	Standard error
W_{forest}	0.643	0.069
B_{forest}	14.68	1.320
A_{forest}	0.279	0.846
W_{scrub}	0.739	0.084
B_{scrub}	10.37	0.940
A_{scrub}	− 0.240	1.350

Table 4. Analysis of variance of soil organic carbon transformed to square roots

Source	Degrees of freedom	Mean square	<i>F</i>	<i>P</i>
Depth	2	5.60	20.70	< 0.001
Vegetation	1	24.13	89.10	< 0.001
Depth × Vegetation	1	1.14	4.22	0.042
Separate non-linear <i>W</i>	1	0.01	0.02	0.888
Residual	150	0.28		
Total	155	9.45		

Parameter estimates and their standard errors

Parameter	Estimate	Standard error
W_{forest}	0.702	0.166
B_{forest}	1.755	0.312
A_{forest}	1.919	0.354
W_{scrub}	0.749	0.281
B_{scrub}	0.923	0.292
A_{scrub}	1.329	0.434

Table 5. Pearson correlation coefficients between SQRT($^{210}\text{Pb}_{\text{ex}}$) activity with SQRT(SOC), stone content and density of the fine fraction at the surface interval (0–2 cm) and for the whole profile (0–14 cm) in forest and scrub soil

	Total		Forest		Scrub	
	SQRT($^{210}\text{Pb}_{\text{ex}}$) /Bq kg ⁻¹		SQRT($^{210}\text{Pb}_{\text{ex}}$) /Bq kg ⁻¹		SQRT($^{210}\text{Pb}_{\text{ex}}$) /Bq kg ⁻¹	
Depth / cm	0–2	0–14	0–2	0–14	0–2	0–14
<i>n</i>	23	156	10	67	13	89
SQRT(SOC) /%	0.758	0.539	0.885	0.683	0.526	0.490
Stone content /%	0.648	0.341	0.481	0.335	0.648	0.331
Density /g cm ⁻³	–0.675	–0.474	–0.939	–0.462	–0.523	–0.527

Boldface numbers are significant at the 95% confidence level

n, total number of soil samples

Table S1. Analysis of variance of $^{210}\text{Pb}_{\text{ex}}$ (Bq kg^{-1}) activity transformed to square roots for all steps in the analysis. Step 4 corresponds with Table 3.

Model parameters and fitting functions for the distribution of $^{210}\text{Pb}_{\text{ex}}$ (Bq kg^{-1}) with an exponential curve that has the following equation:

$$y = \alpha + \beta \times \omega^x,$$

where y is $^{210}\text{Pb}_{\text{ex}}$ activity, x is depth in cm, A represents α , B represents β and W represents ω for each type of vegetation (forest or scrub soil).

STEP 1: Excludes the grouping factor vegetation cover

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	2	481.2	146.8	< 0.001	<i>W</i>	0.686	0.055
Residual	153	3.28			<i>B</i>	12.09	0.745
Total	155	9.45			<i>A</i>	0.132	0.728

Accumulated analysis of variance				
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	481.2	146.8	< 0.001
Residual	153	3.28		
Total	155	9.45		

d.f., degrees of freedom; s.e., standard error

STEP 2: Taking constant parameters separately (Adding separate A_{forest} and A_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	3	323.4	99.56	< 0.001	<i>W</i>	0.686	0.039
Residual	152	3.25			<i>B</i>	12.08	
Total	155	9.45			A_{forest}	0.404	
					A_{scrub}	-0.049	

Accumulated analysis of variance

Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	481.2	148.1	< 0.001
Vegetation	1	7.85	2.41	0.122
Residual	152	3.25		
Total	155	9.45		

d.f., degrees of freedom; s.e., standard error

STEP 3: Taking all linear parameters separately (Adding separate B_{forest} and B_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	4	248.8	80.13	< 0.001	<i>W</i>	0.680	0.038
Residual	151	3.11			B_{forest}	14.35	
Total	155	9.45			A_{forest}	-0.181	
					B_{scrub}	10.41	
					A_{scrub}	0.510	

Accumulated analysis of variance

Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	481.2	154.99	< 0.001
Vegetation	1	7.85	2.53	0.114
Depth × Vegetation	1	24.93	8.03	0.005
Residual	151	3.11		
Total	155	9.45		

d.f., degrees of freedom; s.e., standard error

STEP 4: Taking all parameters separately (Adding separate W_{forest} and W_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	5	199.5	64.16	< 0.001	Wforest	0.643	0.069
Residual	150	3.11			Bforest	14.68	1.320
Total	155	9.45			Aforest	0.279	0.846
					Wscrub	0.739	0.084
					Bscrub	10.37	0.940
					Ascrub	-0.240	1.350

Accumulated analysis of variance				
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	481.2	155.0	< 0.001
Vegetation	1	7.85	2.53	0.114
Depth × Vegetation	1	24.93	8.03	0.005
Separate non-linear W	1	2.36	0.76	0.385
Residual	150	3.11		
Total	150	9.45		

d.f., degrees of freedom; s.e., standard error

Table S2. Analysis of variance of percentage soil organic carbon transformed to square roots for all the steps in the analysis. Step 4 corresponds with Table 4.

Model parameters and fitting functions for the distribution of soil organic C (%) with an exponential curve that has the following equation:

$$y = \alpha + \beta \times \omega^x,$$

where y is $^{210}\text{Pb}_{\text{ex}}$ activity, x is depth in cm, A represents α , B represents β and W represents ω for each type of vegetation (forest or scrub soil)

STEP 1: Excludes the grouping factor vegetation cover

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	2	5.599	13.00	< 0.001	<i>W</i>	0.733	0.188
Residual	153	0.431			<i>B</i>	1.298	0.259
Total	155	0.497			<i>A</i>	1.559	0.364
Accumulated analysis of variance							
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>			
Depth	2	5.60	13.00	< 0.001			
Residual	153	0.43					
Total	155	0.50					

d.f., degrees of freedom; s.e., standard error

STEP 2: Taking constant parameters separately (Adding separate A_{forest} and A_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	3	11.78	42.86	< 0.001	<i>W</i>	0.718	0.111
Residual	152	0.28			<i>B</i>	1.275	
Total	155	0.50			A_{forest}	2.045	
					A_{scrub}	1.251	
Accumulated analysis of variance							
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>			
Depth	2	5.60	20.38	< 0.001			
Vegetation	1	24.13	87.82	< 0.001			
Residual	152	0.28					
Total	155	0.50					

d.f., degrees of freedom; s.e., standard error

STEP 3: Taking all linear parameters separately (Adding separate B_{forest} and B_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	4	9.12	33.89	< 0.001	W	0.714	0.105
Residual	151	0.27			B_{forest}	1.752	
Total	155	0.50			A_{forest}	1.892	
					B_{scrub}	0.916	
					A_{scrub}	1.374	

Accumulated analysis of variance

Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	5.60	20.82	< 0.001
Vegetation	1	24.13	89.69	< 0.001
Depth $\times$ Vegetation	1	1.14	4.24	0.041
Residual	151	0.27		
Total	155	0.50		

d.f., degrees of freedom; s.e., standard error

STEP 4: Taking all parameters separately (Adding separate W_{forest} and W_{scrub})

Summary of analysis					Model parameters		
Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>	Parameter	Estimate	s.e.
Regression	5	7.30	26.94	< 0.001	W_{forest}	0.702	0.166
Residual	150	0.27			B_{forest}	1.755	0.312
Total	155	0.50			A_{forest}	1.919	0.354
					W_{scrub}	0.749	0.281
					B_{scrub}	0.923	0.292
					A_{scrub}	1.329	0.434

Accumulated analysis of variance

Source	d.f.	Mean square	<i>F</i> ratio	<i>P</i>
Depth	2	5.60	20.68	< 0.001
Vegetation	1	24.13	89.11	< 0.001
Depth $\times$ Vegetation	1	1.14	4.22	0.042
Separate non-linear W	1	0.01	0.02	0.888
Residual	150	0.28		
Total	150	0.50		

d.f., degrees of freedom; s.e., standard error