Spatial distribution of natural and artificial radionuclides at the catchment scale (South Central Pyrenees)

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Abstract

Natural and artificial gamma-emitting radionuclides (^{238}U , ^{226}Ra , ^{232}Th , ^{210}Pb , ^{40}K , and ^{137}Cs) were measured in the soils of a small catchment in the Central Pyrenees, Spain. The study was carried out in a mountainous area that was representative of the Tertiary Flysch landscapes in the Southern Pyrenees. Bulk soil cores (n=77) were collected at the intersections of a 200 x 200-m grid established in the Arnás River Catchment. Mean radioisotope activities (Bq kg^{-1}) were 40 (^{238}U), 27 (^{226}Ra), 35 (^{232}Th), 74 (^{210}Pb), 48 ($^{210}\text{Pb}_{\text{ex}}$), 590 (^{40}K), and 31 (^{137}Cs). The ^{210}Pb and ^{137}Cs exhibited the greatest variability, whereas ^{226}Ra and ^{40}K showed the least spatial variation. The relationships between basic soil properties and radionuclide activities indicate that only the radionuclides, $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs , that are fixed to the fine fraction of the soil are directly correlated with the organic matter content, whereas the natural radionuclides are inversely correlated with the carbonate content.

GIS and geospatial interpolations revealed patterns in the spatial concentrations of radionuclides and indicated important differences in their distributions showing the

different behaviour of natural and fallout-derived radionuclides. The radionuclide spatial patterns were strongly correlated with physiographic features such as gradient, orientation, and vegetation cover of the slopes. Within the catchment, the least vegetated and steepest slopes had the lowest radionuclide activities, which suggest that physical processes such as erosion are primary factors in the mobilization of radionuclides in association with soil particles. The results provide insights into the main factors that have affected the spatial distribution of radionuclides in the soils of the catchment, which improves our knowledge of the behaviour of radionuclides in the environment and can lead to a better comprehension of the factors that affect their mobility within ecosystems.

Keywords: natural and artificial radionuclides; ^{238}U , ^{226}Ra , ^{232}Th , ^{210}Pb , $^{210}\text{Pb}_{\text{ex}}$, ^{40}K , and ^{137}Cs ; gamma emitting; spatial distribution; mountain soils; catchment; physiographic and edaphic factors; Central Spanish Pyrenees.

1. Introduction

Natural radioactivity in the soil is mainly due to the decay of radionuclides from the uranium and thorium series. Beck (1972) found that the decay of those isotopes and ^{40}K accounted for 50-80% of the gamma radiation flux at the soil surface. Radiation from natural sources is a major environmental concern because human exposure to natural radionuclides has increased significantly since the 1960s (Baxter, 1991), and more attention has been given to the abundance and distribution of natural radioisotopes in soils (e.g., Kiss et al. 1988; Navas et al., 2002a).

The mineral composition of the parent material controls the natural radioactivity of soils. In addition, radionuclides are adsorbed onto soil components (organic matter, clays, carbonates, Fe/Mn oxides) and take part in biogeochemical processes. Soils play a major role in the cycling of radionuclides and their physicochemical properties

influence the mobility and bioavailability of these radionuclides in terrestrial ecosystems (Kabata-Pendias & Pendias, 2001). Thus, an understanding of the distribution of radionuclides in soils is essential to many environmental studies (Navas et al., 2007). Furthermore, the knowledge of the radionuclide content of soils is central to the establishment of environmental baselines for various substrates and environments (Jordan et al., 1997).

Data on the distribution and behaviour of long-lived radioisotopes in soils is limited (e.g., de Jong et al., 1994). An understanding of the pathways by which radioactivity reaches humans requires an assessment of the soil properties that affect the abundance and distribution of radionuclides. With this purpose most studies have been at the soil profile scale (Sully et al., 1987; Navas et al., 2002a, 2005a; Fujiyoshi & Sawamura, 2004). However, little information exists on the spatial distribution of radionuclides in Mediterranean landscapes where the behaviour of natural gamma-emitting radionuclides can be approached in an integrated approach based on the soil and the environmental processes operating at the catchment scale. This study is aimed to determine the spatial distribution of the natural radionuclides, ^{238}U , ^{226}Ra , ^{232}Th , ^{40}K , ^{210}Pb , $^{210}\text{Pb}_{\text{ex}}$, as well as artificial ^{137}Cs in a small mountain catchment in the central Spanish Pyrenees. In addition, the homogeneity of parent materials in the study area, which is underlain by alternating marls and clay strata, might facilitate the discrimination of factors other than parent materials in controlling the distribution of radionuclides in the landscape.

2. Materials and methods

2.1 . *Study area*

The Arnás River Catchment (284 ha) in the central part of the Spanish Pyrenees was selected for study because the entire area is underlain by Eocene materials. The concentrations of natural radionuclides in soils are related to the nature of the bedrock; thus, the homogeneity of the parent materials provides a common starting point for identifying the distribution patterns of radionuclides in the landscape. Differences in the processes that formed the soil types on the same bedrock would indicate the role of soil processes in the enrichment and depletion of radionuclides.

The study area in Aragón (Huesca province) is described in detail elsewhere (Navas et al., 2005b, 2008). In the area, the elevation is 910-1341 m a.s.l.. The climate is sub-Mediterranean with an oceanic influence. Mean annual rainfall which is of around 900 mm is uniformly distributed within the catchment. The Arnás River divides the catchment into two distinct physiographic areas. The left bank of the river had a main SW aspect, predominant slope gradients of 30-40% and severe erosion (Navas et al., 2005b). The right bank had a NE aspect and predominant slope gradients of 10-20%. The marked physiographic difference between the sides of the catchment might contribute to a better understanding of the physico-chemical processes involved in the spatial distribution of natural and artificial radionuclides.

On Eocene marls and sandstones, six soil types are developed (FAO, 1989) (Figure 1). The bedrock nearby is primarily calcite (64 %), with smaller amounts of quartz (19 %), clays (9 %), feldspars (5 %), and oxides, and the soil mineralogy exhibits significant increases in clays (27% to 41%) and decreases in calcite (33% to 44%). The soils are alkaline, have an average organic matter content of < 4.5% and a carbonate content that varies between 2% for Kastanozems and 17% for Cambisols. Texture is silty loam and bulk density ranges between 1 and 1.2 g cm⁻³, and apart from the organic upper layer, horizon differentiation is minimal. On the left bank (34% of the catchment)

of the Arnás River, Rendzic Leptosols and Calcaric Regosols predominate on steep southwest-facing slopes, and they are shallow and poorly developed on unconsolidated materials. On the right bank (66% of the catchment), the soils on the gentler northeast-facing slopes are Haplic Kastanozems and Haplic Phaeozems. Those brown soils are deeper (50 - > 75 cm) and better developed than are the soils on the SW-facing slopes.

In the study area, the three main vegetation types were forest, shrub, and pasture (Navas et al., 2008). The vegetation included sub-Mediterranean shrubs (*Buxus sempervirens*, *Genista scorpius*, *Echinopartum horridum*, *Juniperus communis*) on the SW-facing slopes (shrub side), with some bare soil and areas of open “matorral” (Mediterranean scrubland). A much denser vegetation of pine (*Pinus silvestris*) and deciduous trees (*Quercus faginea*, *Populus tremula*, *Populus nigra*) occurred on the NE-facing slopes (forest side). Since the Middle Ages, the lands of the catchment were used to grow cereals in terraced fields. In the first half of the 20th Century, major socio-economic changes in Spain promoted widespread emigration from rural to urban areas, which lead to complete and rapid abandonment of the land, which is now only partially used for grazing.

2.2 Sampling and analysis

The soil was sampled at the intersection points within a 200 x 200-m grid, which provided one soil sample collected about every 4 ha (Figure 1). The sampling grid for the semi-detailed scale of the study was considered appropriate for the collection of representative soil samples and the assessment of the soil because of the lithological homogeneity of the study area. We used a 7.5-cm diameter hand-operated corer to collect 77 bulk soil samples to a depth of 20 cm and samples were stored at 4 °C until

they were analyzed. The depth profiles of ^{137}Cs in the catchment indicated that the first 15 cm contained 76-86% of the ^{137}Cs because the soils were undisturbed and very little of the ^{137}Cs was below 20 cm (Navas et al., 2005b); therefore, it was assumed that the samples here contained most of the total areal activity density of the soil at the sampling point.

In the laboratory, soil samples were air-dried, ground, homogenized, and quartered, before being passed through a 2-mm sieve. Soil properties were analysed following standard techniques (CSIC, 1976) which are described in detail by Navas et al. (2008). To quantify organic matter, we used the Sanerlandt Method and a Mettler Toledo titrimeter and electrode. To analyze the clay, silt, and sand fractions, laser equipment was used. To remove the organic matter, samples were disaggregated chemically using 10% H_2O_2 heated to 80 °C, stirred 24 h to facilitate particle dispersion and subjected to ultrasound during the analyses. pH (1:2.5 soil:water) was measured using a pH-meter and carbonates were measured using a pressure calcimeter.

The methods used in the analysis of radionuclides are described in detail elsewhere (Navas et al., 2005a,b). Radionuclide activity in the soil samples was measured using a Canberra high resolution, low background, hyperpure germanium coaxial gamma detector coupled to an amplifier and multichannel analyser. The detector had a relative efficiency of 30% and a resolution of 1.9 keV (shielded to reduce background), and was calibrated using standard samples that had the same geometry as the measured samples. Subsamples of 50 g were loaded into plastic containers. Count times over 24 h provided an analytical precision of about $\pm 5\text{-}10\%$ at the 95% level of confidence. Activities were expressed as Bq kg^{-1} dry soil.

Gamma emissions of ^{238}U , ^{226}Ra , ^{232}Th , ^{40}K , ^{210}Pb , and ^{137}Cs (in Bq kg^{-1} air-dry soil) were measured in the bulk soil samples. Considering the appropriate corrections

for laboratory background, ^{238}U was determined from the 63-keV line of ^{234}Th , the activity of ^{226}Ra was determined from the 352-keV line of ^{214}Pb (Van Cleef, 1994); ^{210}Pb activity was determined from the 47 keV photopeak, ^{40}K from the 1461 keV photopeak; ^{232}Th was estimated using the 911-keV photopeak of ^{228}Ac , and ^{137}Cs activity was determined from the 661.6 keV photopeak. The ^{210}Pb (half-life = 22.26 yr) is integrated by the 'in situ'-produced fraction from the decay of ^{226}Ra (Appleby & Oldfield, 1992) and the upward diffusion of ^{222}Rn in the atmosphere, which is the source of $^{210}\text{Pb}_{\text{ex}}$. Spectrometric measurements were performed a month after the samples were sealed, which ensured a secular equilibrium between ^{222}Rn and ^{226}Ra . The $^{210}\text{Pb}_{\text{ex}}$ activities were estimated from the difference between the total ^{210}Pb activity and the ^{226}Ra activity.

To represent the spatial distribution of the radionuclides, we used the Spline Interpolator Method, which fits a minimum-curvature surface through the input points, to create the output maps of radionuclide activities. Maps of the isolevels of the radionuclides were derived to represent their spatial distributions within the catchment and were overlaid with the isolevels of the soil properties that correlate with the radionuclides. All of the maps, interpolations, and mathematical operations were performed using ArcView GIS 3.2 and ArcGIS 9.0.

To assess the significance of the effects of physiographic conditions, slope and orientation, on radionuclide activities at the sampling points, i.e., on the forest or the shrub side of the catchment, we used an analysis of variance. To that end, the data were divided into forest and shrub sides, and into low, medium and high slope and α was set to 0.05. Correlation coefficients were used to assess the relationships between soil properties and the radionuclides. Finally, the multiple regressions between radionuclides and soil properties and slope were derived using a step-wise methodology.

3. Results and discussion

3.1. Factors affecting the distribution of the radionuclides

In the soils of the Arnás River Catchment, the radioisotope mass activities (Bq kg^{-1}) were 19.9-60 for ^{238}U , 20.8-34.9 for ^{226}Ra , 23.7-49.4 for ^{232}Th , 446-799 for ^{40}K , 26.7-140 for ^{210}Pb , 1.3-113.4 for $^{210}\text{Pb}_{\text{ex}}$, and 4.4 – 64.7 for ^{137}Cs (Table 1). The background levels of natural gamma radionuclides fell within the range of values observed in the region and elsewhere in Spain (Navas et al., 2002b, 2005a) and were similar to those reported by Litaor (1995) and de Jong et al. (1994) in North America and by Fujiyoshi & Sawamura (2004) in Germany.

Radionuclide activities in the soils differed as a function of physiographic features of the landscape. The mean activities of the radionuclides on the two sides of the catchment (Table 2) indicated that the lowest values of all of the radionuclides were on the shrub side of the catchment. An ANOVA detected significant differences between the two sides of the catchment in the mass activities of ^{238}U , ^{226}Ra , ^{232}Th , ^{40}K , ^{210}Pb and $^{210}\text{Pb}_{\text{ex}}$, and, after two outliers that had very high activity concentrations were excluded from the analysis, the difference between the sides of the catchment were apparent in the concentrations of ^{137}Cs . The two outliers were from sampling points that were on the shrub side at the divide of the catchment. They are relicts of the original forest and concentrate radionuclides because the soils are well preserved and maintain a very high organic matter content.

Previous studies have shown that erosive processes were more intense on the shrub side than they were on the forest side (Navas et al., 2005b). On the shrub side, the slopes are steepest and there is the least amount of vegetation cover by compared to the forest side. In contrast, soil stability was predominant on the forest side, where the slopes were gentler on the north-facing right bank side of the Arnás River. In addition,

the two sides of the catchment exhibited distinct patterns in the recovery of the natural vegetation, which is evident in the map of land use (Figure 1) produced from color orthophotographs (Navas et al., 2008). The map indicated three classes of vegetation cover (forest, shrub, pasture) and provided the basis for calculating the area of each land use in the two sides of the catchment by using ArcView GIS 3.2. Differences in vegetation cover were further related to the differences in the main properties of the soils on the two sides of the catchment. The values of the soil properties (organic carbon and nitrogen, available phosphorus and potassium, carbonates, cation exchange capacity, water retention, and water infiltration) indicated that the quality of the soil was better on the forest side than it was on the shrub side (Navas et al., 2008).

In areas of very homogeneous lithology, correlations between radionuclides are expected to be high (Fairbridge, 1972) because the source of the natural radionuclides in soils is the underlying bedrock. In the catchment, the mass activities of ^{40}K , ^{232}Th , ^{226}Ra , and ^{238}U were positively and strongly correlated (Table 3). Thus, members of different decay series were significantly correlated, which indicates that the natural radionuclides (^{238}U - and ^{232}Th -series nuclides, ^{40}K) had a common source: the mineral components of the soil. The relatively weak correlations of ^{238}U with ^{40}K , ^{232}Th , and ^{226}Ra might have been caused by the relatively high mobility of ^{238}U , which was detected in the soil depth profiles of the area (Navas et al., 2002a). Cowart & Burnett (1994) suggested that ^{238}U can form complex ions in calcareous soils and migrate downward in the soil. In similar soils, ^{238}U was depleted at the soil surface due to soil conditions such as pH and organic matter, which favoured the leaching of uranyl complexes (Navas et al., 2002b). Small changes in pH can cause the precipitation of some soil components, mainly carbonates, which in turn can affect the behaviour of uranyl complexes (Winde, 2001).

The absence of a correlation between members of the uranium decay series and ^{40}K and the atmospherically-derived radionuclides ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ reflect differences in their sources and differences in the mobilization of natural radionuclides and ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$. As expected, ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ were positively and significantly correlated, which reflects the similar behaviour of these radionuclides once they become attached to the fine soil fraction.

The relationships between the mass activities of the radionuclides and the clay content indicate that radionuclide activities tend to increase with increasing clay contents although the correlation is only moderate with ^{40}K , ^{232}Th and ^{226}Ra (Table 4). Apparently, natural radionuclides were most abundant in the fine soil fraction because they associated with clay minerals. Although radioisotopes are known to adsorb to clay surfaces or become fixed within the lattice structure (e.g., Jasinska et al., 1982; Vanden Bygaart & Protz, 1995), in the study area, clay content of most of the samples was about 20% (range = 12-27 %). Therefore, the range of values was not sufficiently wide to produce strong correlations with the radionuclides.

Soil carbonate content was significantly negatively correlated with the abundance of each of the natural radionuclides. Carbonate content ranged from 0.11% to 52% and large differences were found between the two sides of the catchment. On the shrub side, typically, carbonate content was >30%, whereas on the forest side, the values were <10% (Navas et al., 2008). The strong correlations between carbonates and the natural radionuclides mass activity density are related to the wide range of carbonate contents, but also to soil processes of carbonate leaching and precipitation involved in their mobilization, especially of ^{238}U and ^{232}Th (Cowart & Burnett, 1994).

Furthermore, in the forest side of the catchment where carbonate content and pH were low, leaching was favoured especially in rich organic soils and, consequently,

some clays have accumulated in the deepest layers (Navas et al., 2005a). However this is not observed at the shrub side where carbonates are abundant. Such differences might also explain the modest correlations between clay fractions and natural radionuclides. In addition, the lack of a correlation between the amount of organic matter and the mass activities of the natural radionuclides supports the association of these radionuclides with the mineral fraction of the soil.

The ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ were not significantly correlated with clay content, but were significantly positively correlated with organic matter content, which was due to the high organic matter content in the upper soil layers (0-15 cm) that generally coincides with the highest activity of these radionuclides because they remain fixed to the fine soil fractions and have little mobility. Moreover, in soil profiles of the study area it was found that some clays accumulated in the deeper soil layers (Navas et al., 2005a). This occurred as a result of leaching processes and was most evident in the soils that had the highest infiltration rates that in turn had the highest organic matter content.

The step-wise regression analysis indicated that about 40% of the variation of the concentration of atmospherically-derived radionuclides was explained by the content of organic matter, to which they are strongly fixed and, to a less extent, in the case of $^{210}\text{Pb}_{\text{ex}}$, by the carbonate content (Table 5). For all of the natural radionuclides, carbonate content was the main explanatory factor and organic matter content was of lesser importance. For ^{226}Ra and ^{232}Th , and ^{40}K , the regression model explained as much as 70% and 60 % of the variation, respectively. The range in carbonate content differed greatly between the two sides of the catchment and the highest carbonate content on the shrub side paralleled the lowest activities of the natural radionuclides, whereas the opposite was true on the forest side. The model explained about 30% of the variation in ^{238}U , which suggested high complexity in the behaviour of this radionuclide. Carbonate

leaching and precipitation, which are influenced by pH, are important factors controlling the solubility of natural radionuclides, especially ^{232}Th and ^{238}U , which become mobilized (Winde, 2001), can migrate, and become fixed by the organic matter.

Various physico-chemical processes influenced the distribution and activity concentrations of the radionuclides at the catchment scale. The ^{226}Ra and ^{232}Th exhibited very similar spatial patterns (Figure 2), which inferred a common source and similar responses to the soil and environmental processes that affected their distribution. In addition, ^{226}Ra had the highest activities on the forest side and the distribution pattern was quite similar to that of ^{232}Th . In the catchment, the sources of ^{226}Ra , a daughter product of ^{238}U , are alkaline rocks, the Eocene clays and marls of the Flysch formation. The ^{226}Ra activity concentrations were slightly lower than those found in soils that had abundant silica in the subsoil (Fujiyoshi & Sawamura, 2004).

Most of the potassium in soils is fixed within a lattice of silicate minerals and, in the three states of potassium in equilibria (solution, exchangeable, and fixed), ^{40}K is only approximately $10^{-2}\%$ (Fujiyoshi & Sawamura, 2004), and the distribution of ^{40}K can be highly variable. The highest activity concentrations of ^{40}K occurred on the forest side and the lowest concentrations were on the shrub side, in the gullies on the forest side, and along the river. The depleted levels of ^{40}K on the shrub side might have been due to loss of mineral fractions of the topsoil horizons. In the soils of a German forest, the activity concentrations of ^{40}K were positively correlated with soil density, which indicated that most of the potassium was contained within the mineral components of the soil (Fujiyoshi & Sawamura, 2004). Furthermore, because most of the potassium is contained within the mineral components, rather than in the organic fraction, the lower levels on the shrub side were due to the loss of the fine fraction through soil erosion.

3.2. Patterns of the spatial distribution of the radionuclides

In the catchment, ^{238}U was highly variable, the highest activity concentrations were on the forest side but lowest activities were found on both sides. Uranium concentrations in soils are associated to soil organic matter content (Valkovic, 2000) and ^{238}U is the most mobile of the radionuclides measured, thus it might have leached from the soil surface to deeper soil layers, which was favoured by the low pH values and highly organic rich soils on the forest side (Navas et al., 2005a). In that way, the lowest concentrations found on the forest side might have been due to significant leaching on this side, which would have produced a pattern similar to that observed on the shrub side, where the physical processes of soil loss might have caused a depletion in ^{238}U .

In general, the highest values of the natural radionuclides (^{238}U and ^{232}Th series nuclides and ^{40}K) were on the forest side and close to the catchment divide. The lowest activities occurred on the shrub side and in other eroded areas of the catchment such as gullies and at the river headwaters. The spatial distribution of the natural radionuclides reflected the heterogeneity of the soil, but also the differences in soil processes occurring on each side of the catchment. Thus, the distribution of those radionuclides was controlled by physical processes such as soil redistribution (erosion/deposition) and by processes of leaching/sorption in the soil complex.

The $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs concentrations exhibited very similar spatial distributions and the maxima values or the depleted levels of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs were highly coincidental throughout the catchment (Figure 3). The initial concentrations of those radionuclides are affected by climate, topography, surface roughness and land cover. The distribution of atmospherically derived radionuclides $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs depend on the spatial variability of fallout. For large areas $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs fallout is clearly

dependent on rainfall. This rainfall dependence related also with an important altitudinal gradient was found for ^{137}Cs in the central Ebro basin (Navas et al., 2007). Recent studies documented a dependence of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs inventories on altitudes (Porto et al., 2009). However this was not the case in the Arnás catchment, because in spite of its altitudinal gradient (430 m), rainfall does not varies within the 284 has catchment.

Although in our study area, the impact of the Chernobyl accident appears to be negligible, research in areas that were affected by this event showed wide variability and complex spatial patterns in ^{137}Cs inventories (e.g., Higgitt et al., 1993). Thus, Golosov et al. (1999) found that the initial ^{137}Cs Chernobyl input showed systematic spatial variability and Van der Perk et al., (2002) also indicate that overland flow during the deposition of Chernobyl-derived ^{137}Cs caused most of its spatial variation.

After deposition, $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs are affected similarly by environmental processes that distribute them in the landscape (Hien et al., 2002; Zhang et al., 2006). Furthermore, the spatial patterns of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs are similar because both radionuclides behave similarly and sorb strongly to soil clays. Excess ^{210}Pb derived from atmospheric dry- and wet-deposition accumulates in the upper soil layers, and surface enrichment of ^{210}Pb down to a depth of approximately 10 cm is common in a variety of soils worldwide (Fowler et., 1998). Like ^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$ becomes rapidly adsorbed onto humic substances and the fine soil fraction, where they remain. Kruyts and Delvaux (2002) suggested that the retention of ^{137}Cs occurs at a very small number of sorbing sites known as the Frayed Edge Sites or weathered micaceous minerals. Both radionuclides are mobilized by physical processes, i.e. wind or water erosion, that are most commonly associated with the fine soil fractions, including some vertical migration and mixing caused by bioturbation can occur (Dorr, 1995). This particular behaviour of the atmospherically derived radionuclides make possible coupling

estimates based on calibrated inventories and geostatistics methods to predict soil redistribution rates and sediments budgets by using ^{137}Cs derived estimates under wind (Chappel, 1998; Chappel and Warren, 2003) and water erosion (Di Stefano et al., 2005; Mabit and Bernard, 2007, Mabit et al., 2008).

The means of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs show significant differences in function of the slope steepness (Table 6). Contrary to the expected the sampling points that were near the river with the lowest slope gradients have the lowest radionuclide concentrations. This is probably because they were affected by bank erosion. When these points were treated as outliers and eliminated from the step-wise analysis (Table 5), slope was a second significant explanatory factor, and together with organic matter content explained 47 % of the variation of ^{137}Cs . The $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs mean concentrations were lower for steep slopes than for gentle slopes, which suggests that the intensity of soil erosion was higher on the steepest slopes and resulted in the depletion of the radionuclides. Although the mean concentrations of natural radionuclides were lower on steep slopes than on gentle slopes, the differences were not statistically significant, which suggests that the effects of physical processes on their distribution are not as important as they are to the distribution of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs radionuclides.

Areas of high erosion were more common on the shrub side of the catchment (Navas et al., 2005b). Water-driven erosion was a very active process, particularly on the convexity of the slopes where there was an abundance of rills. Further evidence of intense soil erosion are the depleted levels of radionuclides on the steepest slopes of the shrub side relative to the high mass activities on the gentler slopes on the forest side. The depleted levels of radionuclides that predominate on the shrub side reflect the effects of runoff acting on the abundant bare surfaces on the shrub side, whereas on the forest side of the catchment, vegetation cover helped to protect soil surfaces (Figures 2

and 3). Evidently, runoff is a key factor in soil erosion, which is a primary force in soil redistribution in the catchment.

Based on the relationships found between soil properties and the radionuclides, we used clay and organic matter content to link their spatial distributions to that of the radionuclides. The overlay of the isolevels of organic matter on that of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ revealed a strong coincidence in their spatial distributions (Figure 3). Thus, atmospherically-derived radionuclides closely agreed with the distribution of organic matter across the catchment. Those radionuclides remain strongly fixed to the fine soil fractions and organic matter; thus, their maxima were highly coincidental with the organic matter maxima. Moreover, the distribution of the lowest mass activities were also coincidental with the distribution of the isolevels of lowest organic matter content, which was most abundant on the shrub side where soil loss was most intense and, consequently, there was a loss of soil organic components and fine mineral soil particles.

Clay isolevels, however, coincided with the distribution of the natural radionuclides (^{238}U and ^{232}Th series nuclides) and ^{40}K (Figure 2). The close agreement between the distributions of the natural radionuclides and clay was particularly noticeable at the sites that had the highest mass activities, which reflected the close link between these radionuclides and the soil mineral components. This agrees with findings by other authors that indicate that radioactivity increases as particle size decreases (Megumi et al., 1982; Baeza et al., 1995).

Despite the apparent positive correlations between clay and the natural radionuclides, the sample size might have been insufficient to detect statistical significance. Yet, the superimposition of their spatial distributions on the distribution of clay content supported those trends. The maxima on the forest side indicated that the

soils there were better preserved and more stable. In the study area, clays are more abundant in soils than they are in the parent materials (Navas et al., 2005a); thus, the distribution of the isolevels of clays and the maxima on the forest side provides further evidence that soil conditions were best in this part of the catchment. Additional evidence was the distribution of physico-chemical soil properties (high nutrients and water content on the forest side) and differences in the recovery of the natural vegetation, which was greater on the forest side than it was on the shrub side of the catchment (Navas et al., 2008).

The ^{238}U and ^{232}Th series commonly occur together in nature and, in most natural systems, the $^{232}\text{Th} / ^{238}\text{U}$ mass ratio is relatively constant (Ivanovich, 1994). Activity Ratios (AR) between parent/parent or between progeny pairs can be used to assess maintenance of the initial proportionality between the ^{232}Th and ^{238}U decay series. Furthermore, $^{238}\text{U}/^{226}\text{Ra}$ activity ratios can be used to ascertain equilibrium within the same decay series. If secular equilibrium prevails in the ^{238}U chain, the activity ratios of $^{238}\text{U}/^{226}\text{Ra}$ will be approximately 1; therefore, values other than 1 indicate disequilibrium. Across the catchment, activity ratios varied considerably (Figure 4), but most of the sampling points had $^{238}\text{U}/^{226}\text{Ra}$ ratios that were >1.0 , which indicated a strong disequilibrium in the ^{238}U chain. That might have been due to large differences in the mobility of those radionuclides because differential mobility is a well-known cause of disequilibrium (e.g., Dowdall and O'Dea, 2002).

In the study area, the concentrations of ^{226}Ra , which has very low mobility (Kabata-Pendias & Pendias, 2001) remained quite constant in the soil profiles (Navas et al., 2005a). Divalent radium cations are strongly absorbed by the hydrous oxides of Fe and Mn (Megumi and Mamuro, 1977) and stable complexing of ^{226}Ra can occur in soils (Taskayev et al., 1978). Apparently, difference in the mobility of ^{238}U and ^{226}Ra

explains the disequilibrium in the $^{238}\text{U}/^{226}\text{Ra}$ ratio. Small variations in pH can affect the behaviour of uranyl complexes (Winde, 2001) and ^{238}U can form complex ions and migrate (Coward & Burnett, 1994). Ratios higher than 1 suggest ^{238}U enrichment. In addition, sorption is an important component of the U cycle (Kabata-Pendias & Pendias, 2001) and significant accumulations of U often occur in rich organic soils and frequently are associated with clays because of the affinity of the clay fraction for absorbing U and Th (Megumi and Mamuro, 1977). In some areas of the catchment, however, ratios were < 1 , which indicated ^{238}U depletion (Figure 2). Differences in pH values, contents of organic matter, and the leaching of carbonates across the catchment (Navas et al., 2008) might have contributed to the increased differential mobility of ^{238}U relative to ^{226}Ra .

The $^{232}\text{Th}/^{226}\text{Ra}$ activity ratio (i.e., progeny pair $^{228}\text{Ac}/^{214}\text{Pb}$) can be used to assess the maintenance of the proportionality within the ^{232}Th and ^{238}U decay series, which in most environmental samples is about 1.1 (Evans et al., 1997). In our study, most of the sampling sites had $^{232}\text{Th}/^{226}\text{Ra}$ activity ratios that were >1.1 (Figure 4), although deviations from the original proportionality were less than they were for the $^{238}\text{U}/^{226}\text{Ra}$ ratio. Like ^{238}U , ^{232}Th can be easily mobilized in forms of various complex inorganic cations and organic compounds. In the soil environment the formation of hydrated cations of Th^{4+} are responsible for its solubility at different pH values (Kabata-Pendias & Pendias, 2001). Thus, similar soil processes as the reported for ^{238}U (leaching and sorption) and as suggested by the coincidental spatial distribution of both radioisotopes (Figure 2) have contributed to the differential mobility of ^{232}Th and ^{226}Ra . Deviations were slightly higher in the lower portion of the catchment, where organic matter and clay contents are higher, which might have caused ^{232}Th enrichment. In the Arnás River Catchment, leaching and sorption were the main processes that

caused the deviations from the original equilibrium within the ^{232}Th and ^{238}U decay series.

4. Conclusions

The factors influencing the spatial distribution of natural radionuclides (^{238}U and ^{232}Th series nuclides and ^{40}K) and atmospherically-derived radionuclides are potentially useful in research on radioactivity at the catchment scale. According to the obtained results it is suggested that in the Arnás River Catchment, the correlations between the radionuclide's mass activity density reflect their different sources: soil minerals for the natural ^{238}U - and ^{232}Th - series nuclides and ^{40}K , and deposition for ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$. The lithological homogeneity of the study area allowed for the derivation of information on the processes involved in the mobility of radionuclides in a Mediterranean mountain environment, and the radionuclides differed significantly in their behaviour. The variability was highest for the atmospheric radionuclides, then in descending order ^{238}U , ^{232}Th , ^{40}K and ^{226}Ra . The distributions of the natural radionuclides tend to be associated with the distribution of the clay fractions across the catchment, which suggests a link with the soil mineral components contained in the fine fractions. The abundance of all radionuclides on the forest side of the catchment reflected the better preservation of the soils under stable soil conditions. The mass activities of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ varied considerably, but their spatial distributions appeared to be most closely associated with the organic matter, which reflected their strong fixation in humic substances and their accumulation in the upper soil layers. The predominance of depleted levels on the shrub side of the catchment, where soil erosion was greatest, reflects the loss of the fine fractions of the soil and suggests a relationship with physical processes such as the erosion involved in soil particle movement. The $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{226}\text{Ra}$ activity

ratios as a measure of disequilibrium caused by the differential mobility of the radioisotopes provided insights into the intensity of soil processes in the catchment and can be of considerable value in similar environments.

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FIGURES

- 1.- Location of the Arnás River Catchment a sub-catchment of the Lubierre River Catchment in the Central Spanish Pyrenees.
- 2.- Spatial distribution of mass activities of ^{226}Ra , ^{232}Th , ^{40}K and ^{238}U (Bq kg^{-1}) across the Arnás River Catchment and overlay of the isolevels of clay content (%).
- 3.- Spatial distribution of mass activities of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ (Bq kg^{-1}) across the Arnás River Catchment and overlay of the isolevels of organic matter content (%).
- 4.- Spatial distribution of the activity ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{232}\text{Th}/^{226}\text{Ra}$ across the Arnás River Catchment.

Figure 1

Figure 2

Figure 3

Figure 4

Table 1.- Basic statistics for the natural and fallout radionuclides (Bq kg⁻¹) in the soils of the Arnás river catchment.

	⁴⁰ K	¹³⁷ Cs	²¹⁰ Pb	²¹⁰ Pb _{ex}	²²⁶ Ra	²³² Th	²³⁸ U
	Bq kg ⁻¹						
mean	586.2	30.9	74.0	48.2	26.7	34.6	40.2
error	9.3	1.3	2.2	2.1	0.4	0.7	1.0
sd	80.3	11.3	19.1	17.9	3.3	5.9	8.3
mínimum	446.0	4.4	26.7	1.3	20.8	23.7	19.9
máximum	799.0	64.7	140.0	113.4	34.9	49.4	60.0

Table 2.- Basic statistics for the natural and fallout radionuclides (Bq kg⁻¹) in the soils at the forest and shrub sides of the Arnás river catchment.

	⁴⁰ K		¹³⁷ Cs		²¹⁰ Pb		²¹⁰ Pb _{ex}		²²⁶ Ra		²³² Th		²³⁸ U	
	Bqkg ⁻¹													
	forest	shrub	forest	shrub	forest	shrub	forest	shrub	forest	shrub	forest	shrub	forest	shrub
mean	602.2	560.8	32.4	28.4	78.8	66.4	52.2	42.0	27.7	25.2	36.1	32.1	42.6	36.5
error	13.5	9.2	1.4	2.5	2.7	3.5	2.5	3.4	0.5	0.4	1.0	0.7	1.2	1.3
sd	91.8	49.4	9.5	13.6	18.0	18.7	16.7	18.4	3.6	1.9	6.5	3.9	8.2	7.2
minimum	446.0	490.0	5.1	4.4	27.8	26.7	1.3	2.3	20.8	21.1	23.7	25.0	21.2	19.9
maximum	799.0	685.0	52.0	64.7	140.0	99.6	113.4	76.9	34.9	28.9	49.4	41.9	60.0	51.1

Table 3.- Pearson correlation coefficients between the radionuclides (Bq kg⁻¹) in the soils of the Arnás river catchment.

	⁴⁰ K	¹³⁷ Cs	²¹⁰ Pb	²¹⁰ Pb _{ex}	²²⁶ Ra	²³² Th	²³⁸ U
¹³⁷ Cs	0.143						
²¹⁰ Pb	0.432	0.647					
²¹⁰ Pb _{ex}	0.330	0.675	0.988				
²²⁶ Ra	0.768	0.087	0.468	0.333			
²³² Th	0.862	0.191	0.478	0.367	0.833		
²³⁸ U	0.501	-0.011	0.240	0.161	0.540	0.522	

Table 4.- Pearson correlation coefficients between radionuclides (Bq Kg⁻¹) and main soil properties in the soils of the Arnás river catchment.

	OM %	Clay %	pH	CO ₃ ⁼ %
¹³⁷ Cs	0.642	-0.021	-0.272	-0.398
²¹⁰ Pb _{ex}	0.561	0.029	-0.355	-0.488
²¹⁰ Pb	0.543	0.083	-0.391	-0.581
²²⁶ Ra	0.109	0.330	-0.354	-0.775
²³² Th	0.228	0.441	-0.412	-0.825
²³⁸ U	0.067	0.281	-0.278	-0.496
⁴⁰ K	0.182	0.394	-0.182	-0.750

Table 5. Step-wise multiple regression analysis between the concentrations of the radionuclides (Bq kg⁻¹) and the soil properties (%) and slope (%).

	Step	variable	R ²	p
¹³⁷ Cs	1	OM	0.41	0.0001
	2	slope	0.43	0.1474
	3	clay	0.44	0.2634
	4	CO ₃ ⁼	0.45	0.2608
²¹⁰ Pb _{ex}	1	OM	0.31	0.0001
	2	CO ₃ ⁼	0.37	0.0150
	3	clay	0.39	0.1000
	4	slope	0.40	0.3200
²²⁶ Ra	1	CO ₃ ⁼	0.60	0.0001
	2	OM	0.71	0.0001
	3	clay	0.72	0.3304
	4	slope	0.72	0.4142
²³² Th	1	CO ₃ ⁼	0.68	0.0001
	2	OM	0.73	0.0003
	3	clay	0.74	0.2818
	4	slope	0.74	0.4042
²³⁸ U	1	CO ₃ ⁼	0.24	0.0001
	2	OM	0.29	0.0275
	3	slope	0.30	0.3185
⁴⁰ K	1	CO ₃ ⁼	0.56	0.0001
	2	OM	0.62	0.0017
	3	slope	0.64	0.0412

p ≤ 0.05 significant at the 95% level of confidence

Table 6.- Least square mean concentrations of the radionuclides (Bq kg⁻¹) for the different slope steepness in the Arnás catchment.

Slope	n	²²⁶ Ra		²³² Th		⁴⁰ K		²³⁸ U		¹³⁷ Cs		²¹⁰ Pb _{ex}	
Low (0-10%)	9	26.0	a	31.3	a	530.0	a	41.1	a	25.9	ab	42.6	ab
Medium (10-20%)	34	27.2	a	36.0	a	605.8	b	41.5	a	34.1	b	54.3	b
High (>20%)	34	26.3	a	33.8	a	578.2	ab	38.8	a	28.6	a	43.3	a

Different letters indicate significant differences at the p-level < 0.05.