Vertical and lateral distributions of ^{137}Cs in cultivated and uncultivated soils on Mediterranean hillslopes

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Abstract

The fallout radionuclide ^{137}Cs has been used widely as an environmental tracer in the study of soil redistribution processes. An understanding of the relationships between ^{137}Cs and soil properties and physiographic factors is essential for a sound interpretation of the estimates of soil redistribution derived from ^{137}Cs data. The purpose of this study was to investigate the relationships of ^{137}Cs with main soil properties in cultivated and uncultivated soils located in the northern border of central part of the Ebro basin in order to infer the behavior of ^{137}Cs in representative soils of Mediterranean mountain hillslopes. The depth distribution of ^{137}Cs and the physicochemical properties of the soils were measured in 59 soil profiles along five soil toposequences that differed in orientation, slope gradient, land uses, soil types and lithologies, and were representative of Mediterranean mountain agroecosystems. The ^{137}Cs mass activities and inventories varied widely (between b.d.l. and 38 Bq kg^{-1}, and between 0 and 2633 Bq m^{-2}, respectively). The highest values were found in surface layers of uncultivated Leptosols (mean: 21.9 Bq kg^{-1}, 1052.0 Bq m^{-2}) associated with steep slopes and high SOM content (mean: 8.3 %), and the lowest values (b.d.l.) were found at deep layers of uncultivated and cultivated soils. In uncultivated soils most ^{137}Cs was found in the upper 12 cm with a clear exponential decay with depth. Cultivated soils had longer mixed ^{137}Cs profiles extending to 47 cm. ^{137}Cs inventories were significantly higher in uncultivated soils (mean: 1616.1 Bq m^{-2}) than in cultivated soils (mean: 1174.1 Bq m^{-2}). The ^{137}Cs contents were significantly positively correlated with SOM and stoniness, respectively, but weakly negatively correlated with clay content. Multivariate analyses were used to test the hypothesis that soil properties and physiographic factors influence the distribution of ^{137}Cs. As much as 80 % and over 60 % of the variance of ^{137}Cs was explained by SOM contents in stepwise model and adding land use and depth intervals in GLM models evidencing the strong control of land use. The results of this study improve our understanding of the effects of soil properties and physiographic factors on the behavior of ^{137}Cs in the Spanish south-central Pyrenees.

Keywords: ^{137}Cs; Vertical distribution; Soil properties; Land uses; Mediterranean environments; Multivariate analysis

1 Introduction

Since the 1970s, fallout ^{137}Cs (half-life 30.2 year) has been used worldwide as environmental tracer to study soil movements and redistribution processes (e.g. Walling et al., 2006; Ritchie et al., 2009; Mabit et al., 2008a; Kato et al., 2010; Li et al., 2011; Porto et al., 2011). Cesium-137 is an artificial radionuclide by-product of the thermonuclear weapons testing that occurred from the 1950s until the early 1970s, with a peak in 1963, or nuclear accidents (e.g. Chernobyl 1986, Fukushima 2011). The Iberian Peninsula was not significantly affected by the release from nuclear accidents

(García-León et al., 1993), and only Chernobyl radiocesium was observed on the Spanish coast of the Mediterranean Sea (Molero et al., 1999). The technique assumes that the fallout ^{137}Cs is spatially uniform and depends on the latitude and the mean annual rainfall (Walling and Quine, 1992). Cesium-137 reaches the soil surface through wet and dry deposition, and soils and sediments, especially clay minerals, rapidly and strongly adsorbed it (Tamura, 1964; Staunton et al., 2002). Once adsorbed to the soil, ^{137}Cs is essentially non-exchangeable, and biological and chemical processes move little of the adsorbed ^{137}Cs through the soil profile. The subsequent content of ^{137}Cs in soils and its redistribution throughout the landscape are primarily controlled by its natural decay, soil redistribution processes, and sediment transport. In most soils, the strong binding of ^{137}Cs to fine soil particles results in a very low plant uptake (Staunton et al., 2002).

The horizontal and vertical movements of soil influenced the distribution of ^{137}Cs, and soil properties (e.g. texture, organic matter content (SOM) and pH) and climate, land use, soil types and management practices influence the vertical migration of ^{137}Cs (Forsberg and Strandmark, 2001). In addition, bioturbation might be another important factor (Müller-Lemans and Van Dorp, 1996). An understanding of the distribution of ^{137}Cs according different physicochemical properties that characterize the soil is important for its use as radiotracer. The contents of ^{137}Cs fixed by different grain-size fractions are strongly correlated with the specific surface areas of the fractions, and a significant preferential adsorption of ^{137}Cs by finer soil particles was demonstrated (He and Walling, 1996). Navas et al. (2007) also found variations in the ^{137}Cs content in a range of grain size fractions of different lithologies across an altitudinal gradient in the central Ebro basin.

The organic matter in the soils is important in the adsorption of ^{137}Cs, and the relationship between SOM and ^{137}Cs has been well studied in a variety of environments (e.g., Ritchie and McHenry, 1973; McHenry and Ritchie, 1977; Martinez et al., 2010; Navas et al., 2011). Although some studies did not find that SOM had an effect on the content of ^{137}Cs in soils (Aslani et al., 2003), most found a close relationship between SOM and ^{137}Cs (e.g. Livens and Loveland, 1988; Mabit and Bernard, 1998; Takenaka et al., 1998; Dumat and Staunton, 1999; Walling and He, 1999). In most soil types, ^{137}Cs is less mobile than are other radionuclides because it is fixed selectively in clay minerals, especially illitic minerals. In organic soils, however, ^{137}Cs is usually more mobile (Dumat and Staunton, 1999; Rosén et al., 1999) because of the high cation-exchange capacity of organic matter (Coughtrey and Thorne, 1983). Recently, Ziembik et al. (2009, 2010) presented a linear regression method for assessment of soil physicochemical parameters that influencing on ^{137}Cs accumulation in forest soils, and they demonstrated that organic matter content, pH, the parameters related to sorption properties of mineral parts and mobile ions concentrations can be used to predict the accumulated ^{137}Cs in soil horizons.

In Mediterranean environments, particularly, soil texture and structure, soil stoniness, clay types, and the geochemical properties of the parent material might have significant effects on the amount of ^{137}Cs in soils (Kiss et al., 1988). In gneiss-cultivated soils, bulk density, stoniness, and vegetation coverage have an effect on the distribution of ^{137}Cs (Schoorl et al., 2004 a).

The ^{137}Cs technique is valid in Mediterranean environments that have high stone content, an abundance of shallow soils, and a steep topography (e.g. Navas and Walling, 1992; Quine et al., 1994; Schoorl et al., 2004 b; Navas et al., 2005 a; Sadiki et al., 2007; Navas et al., 2007; Porto et al., 2009; Estrany et al., 2010; Gaspar et al., 2013), although some of those characteristics of Mediterranean soils might hinder the use of the technique (Chappell, 1999). In Mediterranean agroecosystems, the complex mosaic distribution of land uses along the pathway of runoff circulation might complicate further the use of ^{137}Cs as a radiotracer and, therefore, more needs to be known about the distribution of ^{137}Cs in uncultivated and cultivated soils in that region. Furthermore, the behavior of ^{137}Cs in different soils still requires further investigation, which can be accomplished by considering a greater number of sectioned soil profiles rather than bulk samples, along complete soil toposequences.

The overarching objective of this study was to assess the depth distribution and behavior of ^{137}Cs in uncultivated and cultivated soil profiles along representative soil toposequences in a Mediterranean mountain agroecosystem. Specifically, to assess the effects of land use, soil type, lithology, and physiographic factors on the ^{137}Cs content in the soils and to quantify the relationships between ^{137}Cs and physicochemical soil properties such as SOM, texture, carbonate content and pH. That information was used to develop empirical models that use the most easily measured soil parameters to estimate the content of ^{137}Cs in soils.

2 Materials and methods

2.1 Study area

Representative hillslopes of Mediterranean mountain agroescosystems were selected in the endorheic catchment of the Estaña lake (NE Spain) in the central part of the Pre-Pyrenees, close to the northern boundary of the Ebro river basin (Figure 1).

The relief is abrupt with an altitude level between 676 and 896 m a.s.l. and steep slopes with an average gradient of 19 %. The climate is continental Mediterranean with a mean annual rainfall of 595 mm, mainly distributed in spring and autumn, and a dry summer with high intensity rainfall events (López-Vicente et al., 2008). The mean annual temperature is around 12 ºC with thermal inversions during the winter.

The study area consists of Mesozoic and Neogene materials that include gypsiferous marls, dolomites, limestones, ophites, and sparse saline deposits. The Mesozoic materials are outcrops of Keüper and Muschelkalk Facies. Karstic and hydrologic processes partially control the evolution of the landscape. The soils that predominate are Leptosols, Calcisols, Regosols, Gypsisols and Gleysols bordering the lakes (Machín et al., 2008). In general, most Leptosols are found in the upper part of the catchment where bare rock outcrops are also common on the steeper areas.

The land uses are representative of Mediterranean mountain agrosystems. Winter barley is the main crop and occupies most of the gentle surfaces around the lakes. Steeper slopes are covered by dense and open Mediterranean forest and the remainder are dense and sparse scrublands. The abandoned agricultural fields were left fallow in the first part of last century due to important socio-economic changes and at present are covered with mature shrubs.

2.2 Field sampling

Five representative hillslope transects were selected to represent different complete toposequences within the catchment (Figure 1). The transects were established from the catchment divide to the central lake and are characterized by different land uses, soil types, lithologies, slope gradients, ranges of altitude and orientation. Fifty nine soil profiles were collected along the transects separated 50 m apart each other by using a high resolution GPS system. The sampling point T5-13 was located on a thick Muschelkalk outcrop and a soil sample was not collected from this point.

Soil samples were performed using an 8 cm diameter automatic core driller and a hand-operated corer for shallow soils. The steel core tube was driven automatically or manually into the ground to an average depth of 40 cm, or until the bedrock was reached. The core tubes were subsequently extracted manually and sectioned. Depth increment samples were sectioned at 5 cm increments reaching a maximum depth of 72 cm, obtaining a total of 399 interval samples. Due to the abundance of stones that caused difficulties when sectioning the cores, some depth increments were 10 and 15 cm thick and in some cases the increments were less than 5 cm. In some Leptosols the maximum sampling depth did not extend below 10 cm. Details of the distribution and physiographic characteristics of soil profiles are presented in Table 1.

2.3 Sampling treatment and analysis

Samples were air-dried, hand disaggregated and passed through a 2 mm sieve. The coarse fraction above 2 mm (CF) corresponds to rock fragments and stoniness and was separated from the fine fraction under 2 mm that was used for ^{137}Cs and soil properties analysis.

Several general soil properties [texture, soil organic matter (SOM), pH, electrical conductivity (EC), carbonates ($CO_3^{=}$) and gypsum content] were determined following standard techniques (CSIC, 1976). Grain size of different fractions was analyzed by Coulter laser granulometer after destruction of organic matter with 10 % H_2O_2 at 80 ºC, stirred 24 h to facilitate particle dispersion and subjected to ultrasound during the analyses. The SOM content was determined by a Mettler Toledo titrimeter and electrode. The pH (solid-liquid ratio 1:2.5) was measured using pH-meter, EC (dS m^{-1}) (1,5 soil,water) was measured using a conductivity meter, and carbonates were analyzed using a calcimeter. The gypsum content was determined by the gravimetric method.

The ^{137}Cs activities were measured using high resolution, low background, low energy, hyperpure coaxial gamma-ray detector coupled to an amplifier and multichannel analyser. The detector had an efficiency of 30 % and a 1.9 keV resolution (shielded to reduce background), and was calibrated using standard certified samples in the same geometry as the measured samples. Gamma emission of ^{137}Cs at 661.6 keV was counted for 30,000 s, yielding results with an analytical precision of ± 6-8 % at the 95 % level of confidence. The detection limit in Bq kg^{-1} for ^{137}Cs was 0.2 Bq kg^{-1}. The content of ^{137}Cs in the soil samples is expressed as a concentration or mass activity (Bq kg^{-1}) and as activity per unit area or inventory (Bq m^{-2}) which is calculated using the weight of the < 2 mm fraction and the cross section of the sample.

2.4 Statistical analyses

Correlation coefficients and regressions analyses were used to examine the relationships between ^{137}Cs and soil properties. Mean tests and ANOVA analyses were performed to confirm significant differences between mean mass activities and inventories of ^{137}Cs in terms of physiographic factors, land uses, soil types, vegetation and lithology. Means which differed significantly ($p < 0.05$) were then determined using the Least Significant Difference (LSD Fisher) test.

Furthermore, a principal component analysis (PCA) was used to relate ^{137}Cs content in soils with measured soil properties and physiographic factors. A PCA reduces dimensionality by revealing several underlying components, known as principal components (PC), which are defined as a linear combination of the original variables. The table of component weights shows the estimated value of the original variable for each component extracted, with both positive and negative values. This implies that component weights not only have a greater or lesser signal contributing to the composite signal, but that a signal can also have a negative contribution.

Finally, General Linear Models (GLM) have been developed to predict the content of ^{137}Cs in soils, using significant soil properties more easily measurable (e.g. SOM), and categorical factors (e.g. land use, depth interval, soil type and altitude) as explanatory variables. GLM models unlike standard regression models, allow incorporating both categorical and quantitative factors in the regression analysis. The basic concept of GLM is that the relationship between the dependent variables and the independent variables is expressed as an equation that contains a term for the weighted sum of the values of the independent variables, plus an error term for unexplained effects. Hence, the GLM allows identification of factors that have a significant effect on the response, and how much of the variability in the response variable is attributable to each factor.

3 Results and discussion

3.1 Vertical distribution of ^{137}Cs

The ^{137}Cs mass activities and inventories in the soil profiles (n=59) varied widely, and, the values of the interval samples (n=399) ranged between b.d.l. to 83.1 Bq kg^{-1} and from 0 to 4029.0 Bq m^{-2}, respectively. The high variation in the mass activities and inventories of ^{137}Cs reflected the variation in its depth distribution and the status of the soil i.e. eroding or deposition site (Figures 2.a, 2.b). Other factors such as the distribution of vegetation, stoniness, topographic roughness, and land use can cause wide variations in ^{137}Cs content, as it has been widely recognized in other Mediterranean environments (e.g. Quine et al., 1994; Schoorl et al., 2004a, b; Navas et al., 2007).

The depth distribution of ^{137}Cs mass activities along the toposequences is typical of those found in other uncultivated and cultivated soils (e.g. Walling and Quine, 1995). The exponential decline of ^{137}Cs with depth in the profiles was typical of uncultivated areas, with most of the radioisotope at the top of the soil profile. In cultivated areas, ^{137}Cs was distributed uniformly within the plough layer (Figures 2.a, 2.b).

Anthropogenic disturbance affected profile T4-3 and, in the deepest layers of profile T5-24, which was from a lake sediment deposit, the ^{137}Cs mass activities were extremely high; therefore, these profiles were excluded from the statistical analyses. In the interval samples (n=380) from uncultivated and cultivated soils, the mean mass activities and inventories of ^{137}Cs differed significantly (Table 2). Uncultivated soils had high variability and the highest ^{137}Cs activities and inventories, which were 79 % and 78 % lower, respectively, in the cultivated soils.

The 57 ^{137}Cs profiles from the transects exhibited distinctive features that reflected the diversity of land uses. The depth distribution of the ^{137}Cs mass activities and inventories were typical for the types of land uses (Figure 3). In uncultivated soils, the ^{137}Cs profiles exhibited a typical distribution of this radionuclide, with the highest concentration in the topsoil and a sharp decay with depth, which reflects the adsorption of the fallout ^{137}Cs by the surface soil and limited post-fallout redistribution within the profile. Most (> 80 %) of the ^{137}Cs was in the upper 12 cm of the soil, a peak of ^{137}Cs occurs within the first 6 cm and, generally, no significant ^{137}Cs mass activities were detected below 22 cm. In cultivated soils, however, the radionuclide was thoroughly mixed and relatively uniformly distributed within the plough layer (0-20 cm); however, below this depth, the ^{137}Cs content declined exponentially (Figure 3). The depth to which 80 % of the ^{137}Cs was held in the cultivated soils (21 cm) was almost double the equivalent depth in the uncultivated soils, and the peak of ^{137}Cs increased by 50 % for the cultivated sites (9 cm), and the depth from which ^{137}Cs is not detected increased to 29 cm. These trends are in agreement with observation in other dry and semiarid environments (e.g. Navas and Walling, 1992; Martinez et al., 2010).

3.2 Physicochemical characterization of soils

In the study area the soils were alkaline (mean pH = 8.3) and the highest pH (9.2) was found in calcareous soils on Muschelkalk facies. The soils were stony (mean coarse fraction (> 2 mm) = 32 %), which is typical of Mediterranean soils, that were high as 90 % in some areas of the study site (Table 3). Most (79 %) of the interval samples had a silty-loam texture, although some had silty-clay-loam (13 %), loam (3 %), or sandy-loam (2 %) textures. The mean value of the clay fraction was 23 %, although the samples varied widely (0-83 %). The silt fraction was predominant (mean = 69 %) and the sand fraction was the less abundant (mean = 8 %). The mean SOM was 2.8 % (range = 0.2-19.4 %). The highest SOM contents were found in the shallow surface horizons of forest and dense scrublands but, in cultivated soils the highest content of SOM was 7.8 %. Carbonate content was highest in cultivated Calcisols on the Muschelkalk facies, and the highest gypsum content was found in the soils that developed on Keüper facies. Salinity was highest and pH was lowest in the Gypsisols that surrounded the lake.

The uncultivated and cultivated soils differed significantly in their contents of SOM, clay, CF, carbonate contents, pH and CE. The uncultivated soils had higher contents of SOM, carbonates, and CF, and lower contents of gypsum than did the cultivated soils. The natural vegetation cover and the predominance of limestones as the parent materials of the uncultivated soils influenced those differences. The cultivated soils had the highest contents of the clay fraction, which is characteristic of the Gypsisols that often are used for growing cereal crops (Table 3).

The large variability of the soil properties responds to the heterogeneity of soil types, land uses and vegetation cover in the toposequences, with most of the cultivated fields on the plains and the lower portions of the hillslopes. In addition, vertical differences in soil properties contributed to the high variability in the soil properties with depth (Figure 4). In both cultivated and uncultivated soils, the amount of CF was significantly higher in the upper layers than it was in below. The clay, silt and sand fractions were distributed uniformly with not significant differences between the depth intervals. In uncultivated soils, SOM was highest at the soil surface and decreased exponentially with depth but, in cultivated soils, SOM was distributed homogeneously throughout the soil profile. In uncultivated soils, particularly, pH and carbonate content tended to be highest in the deepest layers which might be caused by the leaching of carbonates, as has been observed in other Mediterranean mountain soils (Navas et al., 2005 b). In the uncultivated soils, EC and gypsum content were similar throughout the profile but, in cultivated soils, these properties varied with depth and were highest in the deepest layers.

The variability in soil properties reflect the differences between soil types that have developed on a variety of lithologies but, particularly, the effects of land use, which is the main reason of the low SOM and CF in cultivated soils.

3.3 Soil properties and other factors that affect the distribution of ^{137}Cs

Land use, soil type, lithology, and altitude gradients affected significantly the mass activities and inventories of ^{137}Cs; however, slope gradient affected inventories, only, and orientation did not have a significant effect on mass activities or inventories. The ^{137}Cs mass activities and inventories were significantly ($p \leq 0.05$) higher in the uncultivated soils than they were in the cultivated soils, which is similar to the differences between forestland and scrubland. The mass activities and inventories of ^{137}Cs were highest in Leptosols, intermediate in the Calcisols and Regosols, and lowest in Gypsisols and Gleysols. The ^{137}Cs mass activities and inventories were significantly lower in the soils on Keüper facies than they were in the soils on Muschelkalk facies. A positive correlation between ^{137}Cs mass activities and inventories and altitude paralleled an increase in forestland. The ^{137}Cs inventories, but not the mass activities were significantly higher on the steepest slopes. Apparently, ^{137}Cs was strongly affected by land use and, therefore, the interval samples from the two types of soils were evaluated separately to assess the effects of the rest of the factors (Figure 5). ^{137}Cs activities and inventories were significantly higher in uncultivated soils that were covered by forest than they were in scrubland areas, and significantly higher in areas between 700 and 900 m than they were at lower elevations. The lithology had an effect on ^{137}Cs inventories, only, which were highest in a colluvial deposit. In cultivated soils, the significantly higher ^{137}Cs mass activities and inventories were found in doline deposits and Keüper facies, respectively, and in Regosols. ^{137}Cs mass activities were highest on north-facing sites, and inventories were higher in flat areas than they were on cultivated steeper slopes.

The mass activities of ^{137}Cs were significantly positively correlated with SOM and CF, and negatively correlated with clay and pH, and, with the exception of CF, the same relationships applied to the inventories (Table 4), although the strength of the correlations usually was greater for ^{137}Cs activities than they were for ^{137}Cs inventories (see also Martinez et al. (2010)).

The strength of the correlations between ^{137}Cs activities and inventories and soil properties was significantly higher in the uncultivated soils, where ^{137}Cs activities and inventories were positively correlated with SOM, CF, EC, and gypsum content, and negatively correlated with carbonates and pH. In cultivated soils, SOM was the only soil property that was significantly positively correlated with ^{137}Cs activities and inventories. Furthermore, ^{137}Cs mass activities were inversely correlated with clay, EC and gypsum, and ^{137}Cs inventories were inversely correlated with the pH. All of the transects, except T1, exhibited similar relationships, although with slight increases of significance of the correlation coefficients. Probably, the characteristics of the soils and the lithology of the substrate were the underlying causes of the significant negative correlations between ^{137}Cs and carbonates, pH, and EC.

In uncultivated soils, ^{137}Cs activity and CF content were positively correlated, which has been observed elsewhere (Lu and Higgitt, 2000; Schoorl et al,. 2004 a) and its reflects the rapid adsorption of ^{137}Cs within the soil and of its limited vertical movement. Assuming the same amount of ^{137}Cs deposited as in other surfaces, the ^{137}Cs adsorbed per unit mass of soil matrix becomes higher when less soil matrix for adsorption is available.

Radioisotopes are adsorbed onto clay surfaces or fixed within the lattice structure (e.g., Sawhney, 1972; Maes and Cremers, 1986; Cornell, 1993; He and Walling 1996; Wallbrink and Murray, 1996) and the adsorption is highly specific, particularly on illitic materials, which are thought to contain a small proportion of sites, frayed edge sites (FES), which have a very strong affinity for ^{137}Cs (Brouwer et al., 1983). However, in this study ^{137}Cs was not significantly correlated with the overall clay sized faction. The highly homogenous depth distribution of clay in the uncultivated and cultivated soils, and the limited range of variation in the clay fraction (85 % of the samples had clay content between 15 and 30 %) might explain the lack of a significant correlation between clay content and ^{137}Cs. The SOM of soils is important to the adsorption of ^{137}Cs, but this adsorption is supposed to be non-specific, and is influenced by the cation exchange capacity (CEC) of the organic matter, compared with the specific adsorption of ^{137}Cs on clays (Rigol et al., 2002). Dumat and Staunton (1999) suggest that organic matter is preferentially adsorbed on FES, and SOM decreases the adsorption of ^{137}Cs in clays and might inhibit the adsorption of ^{137}Cs on illite and montmorillonite, particularly, on illite when the ^{137}Cs concentration level is very low (Kim et al., 2006).

To eliminate the collinearity between properties that have similar depth distribution patterns, the relationships between ^{137}Cs and SOM and clay fraction were evaluated for each depth interval, individually. The correlations between ^{137}Cs and SOM were stronger at the upper depth intervals, particularly, in uncultivated soils, and, in the cultivated soils, the correlations were not statistically significant (Table 5). At depths below 15 cm, the correlations were weaker and were non-significant at the lowest depths. At the 10-15 cm interval, the absence of a significant correlation between ^{137}Cs and SOM suggests that, in the absence of high SOM, ^{137}Cs (particularly, inventories) is positively correlated with clay content (Dumat et al., 2000; Staunton et al., 2002).

A Principal Components Analysis (PCA) indicated that combinations of variables explained a relatively high proportion of the total variation between the samples. Four components were retained that had eigenvalues > 1 and explained 74 % of the total variance. The first component accounted for 27 % of the total variance and had high values for the variables associated with the physiographic factor (altitude and slope gradient), CF, ^{137}Cs mass activity, and SOM. The first component reflects the high ^{137}Cs mass activities in the uncultivated Leptosols and Calcisols on steep forest slopes that had high SOM and CF content and the low ^{137}Cs mass activities in the cultivated soils on gentle slopes, where SOM and CF content were low, and the fine fraction was relatively large. The second component, which explained 19 % of the total variance, was associated with grain size and identified negative correlations between clay and silt fractions with sand fractions. The third component, which explained 17 % of the variance, reflected the carbonate content and slope gradient. The fourth component, which explained 11 % of the total variance, identified high loading values of altitude, slope gradients and clay fraction that were negatively correlated with CF and silt fraction that was associated with the profiles on the cultivated lowlands (Table 6 and Figure 6). Similar results were obtained when ^{137}Cs inventories were used instead of ^{137}Cs activities. In both cases, the weights of the variables were distributed among several components and the results were affected by the depth distribution of soil properties and the position of the soil profile along the toposequence. Although mathematically, PCA can not be used to make predictions, the PCA biplot test the significance of the correlations between the analyzed variables in all of the interval samples, and the results are consistent with the conclusions drawn from the correlation analysis.

3.4 Modeling ^{137}Cs as a function of soil properties and physiographic factors

A generalized pattern of soil properties and physiographic factors is observed across the southern part of the Spanish Central Pyrenees. In the region it is usual to find a soil catena developed from the upper part of slopes to the bottom part. Physicochemical soil properties, soil types, vegetation cover and land uses typically differ in soils along the catena. Land uses are highly spatially correlated with hillslope position and have wide ranging effects on soil properties and vegetation cover. Our aim was to gain an understanding of the role played by these main factors and how they are involved in the soil processes operating in the southern part of the Central Pyrenees where local climate, physiographic and edaphic conditions are within the ranges in this study.

3.4.1 Models for soil profiles

^{137}Cs mass activity of the soil profiles was simulated using a step-wise regression model that explained 80 % of the variance at the 95 % confidence level with a standard error of 2.5 (Equation 1). SOM and the depth of the profile, respectively, explained 71 % and 9 % of the total variance in the ^{137}Cs mass activity (Figure 7.a). We tested with different significant factors but a GLM did not improve these results.

$$^{137}Cs\ (Bq\ kg^{-1}) = 9.95 + 0.88\ SOM\ (\%) - 0.19\ depth\ profile\ (cm) \quad (1)$$

Less satisfactory results were obtained when modeling the ^{137}Cs inventory, which others have observed in cultivated soils (de Jong et al., 1986; Mabit et al., 2008b). Compared with the results of a multivariate regression equation, a GLM model fitted better by including the SOM content, depth of the profile, land use, soil type, and altitude. However, equation 2 only explained 30 % of the variance in ^{137}Cs inventories at the 95 % confidence level with a high error, which suggests that there was greater complexity in the behavior of the ^{137}Cs inventory than there was in the activity.

$$^{137}Cs\ (Bq\ m^{-2}) = 1849.56 - 241.92\ I1(1) + 27.02\ I1(2) - 301.58\ I2(1) - 197.63\ I3(1) + 47.17\ I3(2) - 61.57\ SOM\ (\%) - 4.19\ depth\ profile\ (cm) \quad (2)$$

where,

I1(1) = 1 if Soil Type= Calcisols; I1(2) = 1 if Soil Type= Leptosols; for both cases -1 if Soil Type= Regosols and 0 otherwise.

I2(1) = 1 if Land Use=Cultivated, -1=uncultivated, 0 otherwise

I3(1) = 1 if Altitude= 600-700 m; I3(2) = 1 if Altitude= 700-800 m; for both cases -1 if Altitude= 800-900 m and 0 otherwise.

The model proposed underestimated the highest values of ^{137}Cs inventories, and overestimated the lowest values (Figure 7.b); therefore, to increase the amount of the variance explained by the model, a larger number of observations are required.

3.4.2 Models for interval samples

The step-wise regression analysis indicated that SOM content, clay fraction and pH explained about 51 % of the variance in ^{137}Cs mass activities, with a standard error of 3.0 at the 95 % confidence level (equation 3).

$$^{137}Cs\ (Bq\ kg^{-1}) = 24.10 + 1.60\ SOM\ (\%) - 0.09\ Clay\ (\%) - 2.64\ pH \qquad (3)$$

However, clay fraction and pH explained around 1 % and 0.9 % of the variance, respectively. Therefore, the model was simplified and fitted to equation 4 in which SOM appears as the only predictor of the ^{137}Cs mass activities (Figure 8.a).

$$^{137}Cs\ (Bq\ kg^{-1}) = -\,0.27 + 1.70\ SOM\ (\%) \qquad (4)$$

A GLM model that took into account land use and depth interval, the factors that were significantly correlated with ^{137}Cs, produced better results than did the regression analyses. The GLM model explained around 68 % of the variance in the ^{137}Cs mass activities and fitted to equation 5 with a standard error of 3.5 (Figure 8.b).

$$^{137}Cs\ (Bq\ kg^{-1}) = 23.72 + 8.04\ I1(1) + 1.55\ I1(2) + 0.33\ I1(3) - 0.23\ I1(4) - 2.05\ I1(5) - 0.61\ I1(6) - 3.84\ I1(7) - 0.39\ I1(8) - 0.78\ I1(9) - 1.27\ I1(10) - 1.45\ I1(11) + 4.11\ I1(12) - 1.26\ I1(13) - 1.03\ I1(14) - 0.85\ I2(1) + 0.78\ SOM\ (\%) - 2.79\ pH \qquad (5)$$

where,

I1(1) = 1 if Depth Interval= 0-5 cm; I1(2) = 1 if D.I=10-15 cm; I1(3) = 1 if D.I=15-20 cm; I1(4) = 1 if D.I=20-25 cm; I1(5) = 1 if D.I=20-30 cm; I1(6) = 1 if D.I=25-30 cm; I1(7) = 1 if D.I=25-35 cm; I1(8) = 1 if D.I=30-35 cm; I1(9) = 1 if D.I=35-40 cm; I1(10) = 1 if D.I=40-45 cm; I1(11)

= 1 if D.I=45-50 cm; I1(12) = 1 if D.I=5-10 cm; I1(13) = 1 if D.I=50-55 cm; I1(14) = 1 if D.I=55-60 cm; for all cases -1 D.I=60-65 cm and 0 otherwise.

I2(1) = 1 if Land Use=Cultivated, -1=uncultivated, 0 otherwise

Similar results were obtained when the inventories of ^{137}Cs were modeled although the percentages of variance explained by regression models were lower. The step-wise regression model explained about 29 % of the variance in the ^{137}Cs inventories, SOM was the property with the highest explanatory power, while clay fractions explained 2 % of the variance (equation 6). However, the GLM model that included land use and depth intervals (equation 7) increased the percentage of variance and explained about 60 % of variance in the ^{137}Cs inventories (Figure 8.c).

$$^{137}Cs\ (Bq\ m^{-2}) = 155.92 + 62.59\ SOM\ (\%) - 5.04\ Clay\ (\%) \quad (6)$$

$$^{137}Cs\ (Bq\ m^{-2}) = 893.80 + 410.48\ I1(1) + 91.59\ I1(2) + 37.90\ I1(3) - 7.10\ I1(4) - 4.08 I1(5) - 40.18\ I1(6) - 116.76\ I1(7) - 46.47\ I1(8) - 65.85\ I1(9) - 88.74\ I1(10) - 100.90\ I1(11) + 220.14\ I1(12) - 98.34\ I1(13) - 94.17\ I1(14) - 27.20\ I2(1) + 10.48\ SOM\ (\%) - 97.44\ pH \quad (7)$$

where,

I1(1) = 1 if Depth Interval= 0-5 cm; I1(2) = 1 if D.I=10-15 cm; I1(3) = 1 if D.I=15-20 cm; I1(4) = 1 if D.I=20-25 cm; I1(5) = 1 if D.I=20-30 cm; I1(6) = 1 if D.I=25-30 cm; I1(7) = 1 if D.I=25-35 cm; I1(8) = 1 if D.I=30-35 cm; I1(9) = 1 if D.I=35-40 cm; I1(10) = 1 if D.I=40-45 cm; I1(11) = 1 if D.I=45-50 cm; I1(12) = 1 if D.I=5-10 cm; I1(13) = 1 if D.I=50-55 cm; I1(14) = 1 if D.I=55-60 cm; for all cases -1 D.I=60-65 cm and 0 otherwise.

I2(1) = 1 if Land Use=Cultivated, -1=uncultivated, 0 otherwise

The models that were developed for uncultivated soils individually explained a proportion of the variance that was similar to that calculated for the total interval samples. In contrast, in the models for cultivated soils, the proportion of the variance explained was smaller.

In this area, slope aspect, topography, land use, vegetation cover and soil types are interrelated, and determine the physical and chemical properties of soils. In general, uncultivated areas of Mediterranean forest and scrublands are located on the upper part of the steep slopes on stony Leptosols, and Regosols, while cultivated fields of winter barley predominate on more gentle slopes, on Regosols, Calcisols and Gypsisols. Accordingly, there is a clear pattern of decreasing organic matter, stoniness and carbonates, and increasing fine fraction, EC, gypsum content and pH, from the upper uncultivated part to the cultivated bottom part. This generalized pattern is the physical context of the empirical models developed.

Correlation of soil properties with terrain attributes such as slope gradient, curvature and elevation, has been quantified statistically, and quantitative models have been proposed to elucidate the spatial distribution of soil properties in landscape and ecosystem processes (e.g. Yoo et al., 2006; Hancock et al., 2010). The understanding that hillslope morphology and topography modifies water flow and soil redistribution processes and soil patterns in landscapes have been extensively reported (e.g. Gessler et al., 2000). Previous research in the study area also indicated that physical processes of soil erosion and deposition are involved in the redistribution of ^{137}Cs in soils. The fact that SOM was significantly correlated with ^{137}Cs (stepwise regression model, eq. 1), as found in other environments (e.g. Mabit et al., 2008b; Martinez et al., 2010), suggests that vegetation cover and land use have a strong control on the content of ^{137}Cs in soils. Thus, at forest sites with higher slopes compared to cultivated sites, the land cover protects the soil surface against erosion and counteracts the effect of the slope gradient.

The GLM models provided great capacity to predict the mass activities and inventories of ^{137}Cs (eq. 6 and 7, respectively) in interval samples of the study area. If SOM and pH, which are properties that are more easily measured than is ^{137}Cs content, are known, and land use and depth intervals are included in the model, the mass activities and inventories of ^{137}Cs can be predicted satisfactorily. Those models are useful for estimating the ^{137}Cs content within the first 5 cm of the soil, which is of interest in fingerprinting studies. In addition, the models are useful for completing data of ^{137}Cs content in the soil profiles from similar Mediterranean environments, particularly, for the deepest intervals.

The highly heterogeneous agroecosystems, which exhibit high variability in soil properties, management practices, and intricate landscape, contributed to the uncertainty in the predicted values. An increase in the number of observations might increase the amount of variance explained by those models and a real test would be of interest to validate the results of the models, although further research is needed. Our study aims to contribute to reducing the analytical and experimental costs of using fallout ^{137}Cs.

The results provide insight into the main factors that have affected the spatial distribution of ^{137}Cs in the soils of the study area, which improves our knowledge of the behavior of the radionuclide in the Spanish south-central Pyrenees and can lead to a better comprehension of the factors that affect its distribution within ecosystems.

4 Conclusions

In Mediterranean mountain agroecosystems, land use influences the depth distribution in ^{137}Cs and the radioisotope content is significantly positively correlated with SOM content. In the northern border of central part of the Ebro basin, uncultivated and cultivated soils differed significantly in the depth distribution of ^{137}Cs and soil properties. Along the toposequences, the most important soil properties such as SOM and CF appeared to have influenced the variation of ^{137}Cs mass activity, which reflects the differences in the land management practices and soil characteristics within the study area.

The significant correlations found between ^{137}Cs and some soil properties, and an understanding of the factors and processes that significantly affect the radioisotope content represent a promising basis for the development of predictive models of ^{137}Cs content in soils. Our results could form the basis for statistical models of ^{137}Cs content in soils in the study area and even in other Mediterranean agroecosystems environments with similar physiographic and climatic conditions. However, this will require further research and validation in other geographical locations. The development of such predictive equations will reduce the experimental and economic costs of applying fallout ^{137}Cs and our study aims to contribute to this objective.

Knowledge of the behavior and distribution of ^{137}Cs in soils is of importance for understanding how this radionuclide is distributed within the ecosystems and, therefore, for interpreting the information that it provides as soil tracer. The information gained with this study has implications for the use of this radionuclide in future research as a radiotracer in studies of soil erosion and soil redistribution processes and sediment budgets.

This study shed light on the behavior of ^{137}Cs in highly heterogeneous environments such as the intricate mosaic of land uses and soil types found in Mediterranean mountain landscapes.

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Tables

Table 1. Number of sampling points and interval samples for the transects according to edaphic and physiographic characteristics.

	Soil profiles						Interval samples					
	Total	T1	T2	T3	T4	T5	Total	T1	T2	T3	T4	T5
Total	59	4	7	12	13	23	399	22	46	86	102	143
Land use												
Uncultivated	38	_	7	10	6	15	224	_	46	67	40	71
Cultivated	21	4	_	2	7	8	175	22	_	19	62	72
Soil type												
Calcisols	25	_	4	3	9	9	164	_	27	19	68	50
Gleysols	5	1	1	1	1	1	50	7	14	8	11	10
Gypsisol	3	_	_	_	_	3	30	_	_	_	_	30
Leptosols	11	_	_	6	2	3	57	_	_	40	13	4
Regosols	15	3	2	2	1	7	98	15	5	19	10	49
Lithology												
Muschelkalk f.	37	_	5	9	11	12	235	_	41	59	81	54
Keüper f.	16	2	2	2	2	8	134	14	5	19	21	75
Doline deposit	3	2	_	1	_	_	16	8	_	8	_	_
Colluvial depoit	3	_	_	_	_	3	14	_	_	_	_	14
Altitude m												
600-700	26	4	4	7	4	7	181	22	11	51	35	62
700-800	23	_	3	5	9	6	175	_	35	35	67	38
800-900	10	_	_	_	_	10	43	_	_	_	_	43
Slope gradient %												
0-12	26	2	5	6	6	7	188	14	34	44	45	51
12-24	23	2	2	5	6	8	158	8	12	38	49	51
>24	10	_	_	1	1	8	53	_	_	4	8	41

Table 2. Basic statistics of the ^{137}Cs mass activities and inventories for the interval samples.

	^{137}Cs Bq kg^{-1}			^{137}Cs Bq m^{-2}		
	Total	Uncultivated	Cultivated	Total	Uncultivated	Cultivated
n	380	215	165	380	215	165
Mean	4.6	6.2	2.4	219.2	278.1	142.3
Median	2.5	2.4	2.6	124.4	111.3	132.6
SD	6.5	8.0	2.2	323.1	403.7	134.1
CV %	142.5	130.3	90.0	147.4	145.2	94.2
Min.	b.d.l.	b.d.l.	b.d.l.	0.0	0.0	0.0
Máx.	37.9	37.9	8.1	2632.8	2632.8	581.7

SD standard deviation
CV coefficient of variation
b.d.l. below detection limit

Table 3. Basic statistics of the main physicochemical soil properties for the interval samples.

	Clay %	Silt %	Sand %	<2mm %	CF %	SOM %	pH	EC dSm^{-1}	$CO_3^{=}$ %	Gypsum %
Uncultivated n=224										
Mean	21.4	69.3	9.3	61.8	38.2	3.6	8.4	0.2	49.5	5.6
Median	22.0	72.3	4.3	61.3	38.7	2.6	8.4	0.2	50.0	5.1
SD	6.9	12.1	15.3	19.1	19.1	3.1	0.2	0.1	15.2	2.6
CV %	32.4	17.4	164.9	30.9	49.9	86.4	2.8	41.6	30.7	46.6
Min.	0.3	9.5	0.0	17.0	0.0	0.2	7.5	0.1	6.7	1.0
Máx.	77.9	84.9	90.2	99.8	83.0	19.4	8.8	0.6	80.5	17.2
Cultivated n=175										
Mean	24.1	69.1	6.9	77.1	22.9	1.8	8.2	0.8	34.2	7.2
Median	23.4	71.5	2.4	82.1	18.0	1.7	8.3	0.2	33.5	4.5
SD	8.5	11.3	13.4	19.7	19.7	1.2	0.3	1.0	17.6	8.5
CV %	35.2	16.3	195.0	25.5	86.2	63.8	4.1	123.1	51.5	117.6
Min.	0.6	9.9	0.0	19.5	0.0	0.2	7.3	0.1	7.7	0.7
Máx.	83.4	83.0	87.0	99.8	80.5	7.8	9.2	2.7	99.8	51.3

SD standard deviation
CV coefficient of variation
(i.e. CF = coarse fraction, SOM = soil organic matter, EC = electrical conductivity, $CO_3^{=}$ = carbonates)

Table 4. Pearson correlation coefficients between ^{137}Cs mass activities and inventories with main soil properties for the interval samples, differentiating between uncultivated and cultivated soils.

	^{137}Cs Bq kg^{-1}			^{137}Cs Bq m^{-2}		
	Total	Uncultivated	Cultivated	Total	Uncultivated	Cultivated
^{137}Cs Bq kg^{-1}				**0.87**	**0.86**	**0.92**
^{137}Cs Bq m^{-2}	**0.87**	**0.86**	**0.92**			
CF %	**0.21**	**0.16**	0.09	0.04	-0.01	-0.12
Clay %	**-0.15**	-0.11	**-0.15**	**-0.15**	**-0.16**	-0.09
Silt %	-0.03	-0.03	-0.02	0.00	0.00	0.03
Sand %	0.10	0.07	0.12	0.08	0.07	0.03
SOM %	**0.70**	**0.68**	**0.42**	**0.52**	**0.49**	**0.35**
$CO_3^{=}$ %	-0.09	**-0.30**	-0.09	-0.08	**-0.24**	-0.13
pH	**-0.30**	**-0.50**	**-0.20**	**-0.20**	**-0.30**	**-0.23**
EC dSm^{-1}	-0.09	**0.57**	**-0.16**	-0.06	**0.37**	-0.03
Gypsum %	0.06	**0.46**	**-0.15**	0.05	**0.35**	-0.09

Bold face numbers are significant at $p \leq 0.05$
(i.e. CF = coarse fraction, SOM = soil organic matter, EC = electrical conductivity, $CO_3^{=}$ = carbonates)

Table 5. Pearson correlation coefficients between ^{137}Cs mass activities and inventories with SOM and clay fraction for each depth interval.

	0-5	5-10	10-15	15-20 cm	20-25	25-30	30-35	25-40	40-45
				^{137}Cs Bq kg^{-1}					
Uncultivated									
clay %	0.20	**-0.39**	-0.22	0.26	-0.18	0.18	-	-	-
SOM %	**0.32**	**0.39**	**0.60**	0.18	0.23	**0.61**	-	-	-
Cultivated									
clay %	0.00	0.06	-0.23	0.09	-0.31	**-0.49**	**-0.48**	-0.15	0.23
SOM %	0.20	0.33	0.42	0.15	-0.12	0.19	0.28	0.34	0.15
				^{137}Cs Bq m^{-2}					
Uncultivated									
clay %	0.19	**-0.44**	**-0.41**	0.21	-0.16	0.09	-	-	-
SOM %	**-0.37**	-0.05	0.22	0.03	0.07	**0.54**	-	-	-
Cultivated									
clay %	-0.14	0.31	-0.12	0.26	-0.13	**-0.46**	-0.44	-0.18	0.23
SOM %	-0.01	0.38	0.22	0.10	-0.06	0.07	0.26	0.33	0.15

Bold face numbers are significant at $p \leq 0.05$
(i.e. SOM = soil organic matter)

Table 6. Principal component loading (PC_i) for the four first components including a) ^{137}Cs mass activity and b) ^{137}Cs inventories.

n= 380	a)				b)			
	PC_1	PC_2	PC_3	PC_4	PC_1	PC_2	PC_3	PC_4
^{137}Cs	0.26	0.25	-0.53	0.19	0.20	-0.19	-0.56	-0.23
Altitude m	0.34	0.28	-0.01	-0.52	0.35	-0.30	-0.15	0.46
Slope gradient %	0.25	0.31	0.27	-0.47	0.26	-0.36	0.14	0.48
CF %	0.35	0.08	0.05	0.33	0.34	-0.07	0.12	-0.27
Clay %	-0.33	0.26	-0.04	-0.36	-0.34	-0.24	-0.04	0.44
SOM %	0.28	0.32	-0.51	0.08	0.23	-0.26	-0.54	-0.19
EC dSm^{-1}	-0.36	-0.14	-0.33	-0.16	-0.37	0.19	-0.31	0.13
Sand %	0.37	-0.55	-0.13	-0.12	0.38	0.55	-0.12	0.10
Silt %	-0.24	0.51	0.19	0.38	-0.25	-0.52	0.17	-0.41
$CO_3^{=}$ %	0.35	0.01	0.46	0.19	0.37	-0.07	0.46	-0.12

(i.e. CF = coarse fraction, SOM = soil organic matter, EC = electrical conductivity, $CO_3^{=}$ = carbonates)

774 **Figures**

775 Figure 1. The study area in the northern border of central part of the Ebro basin and the sampling sites on five transects.

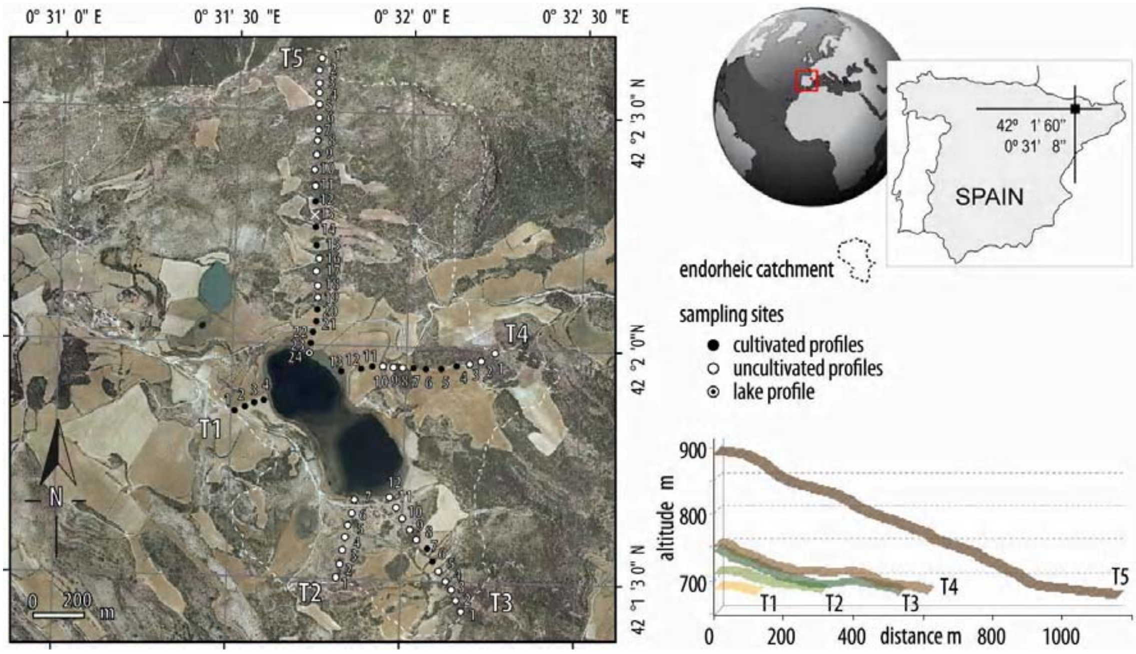

Figure 2.a. Depth distribution profiles for ^{137}Cs mass activity and inventory along transects T1, T2, T3 and T4.

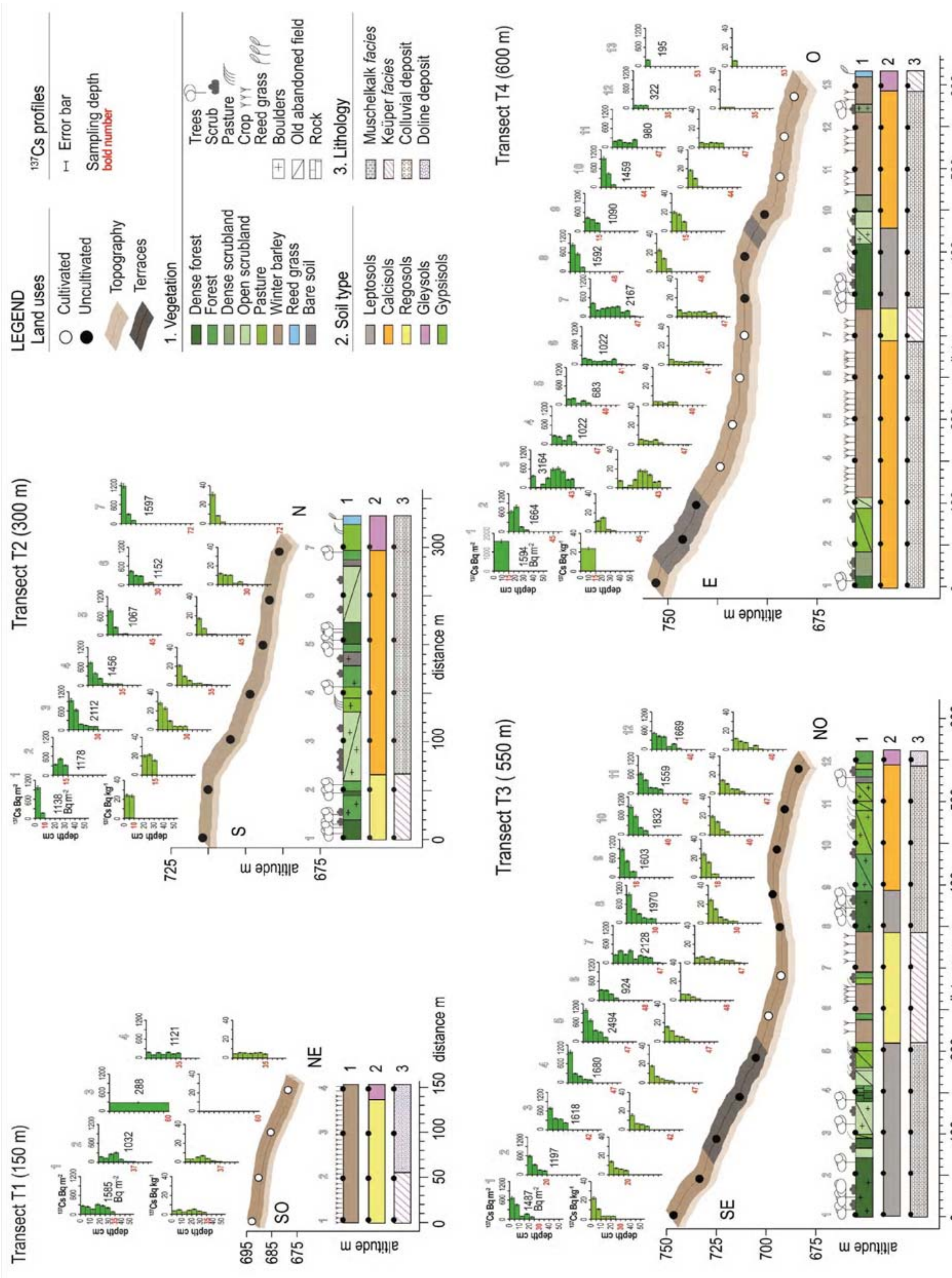

Figure 2.b. Depth distribution profiles for ^{137}Cs mass activity and inventory along transect T5.

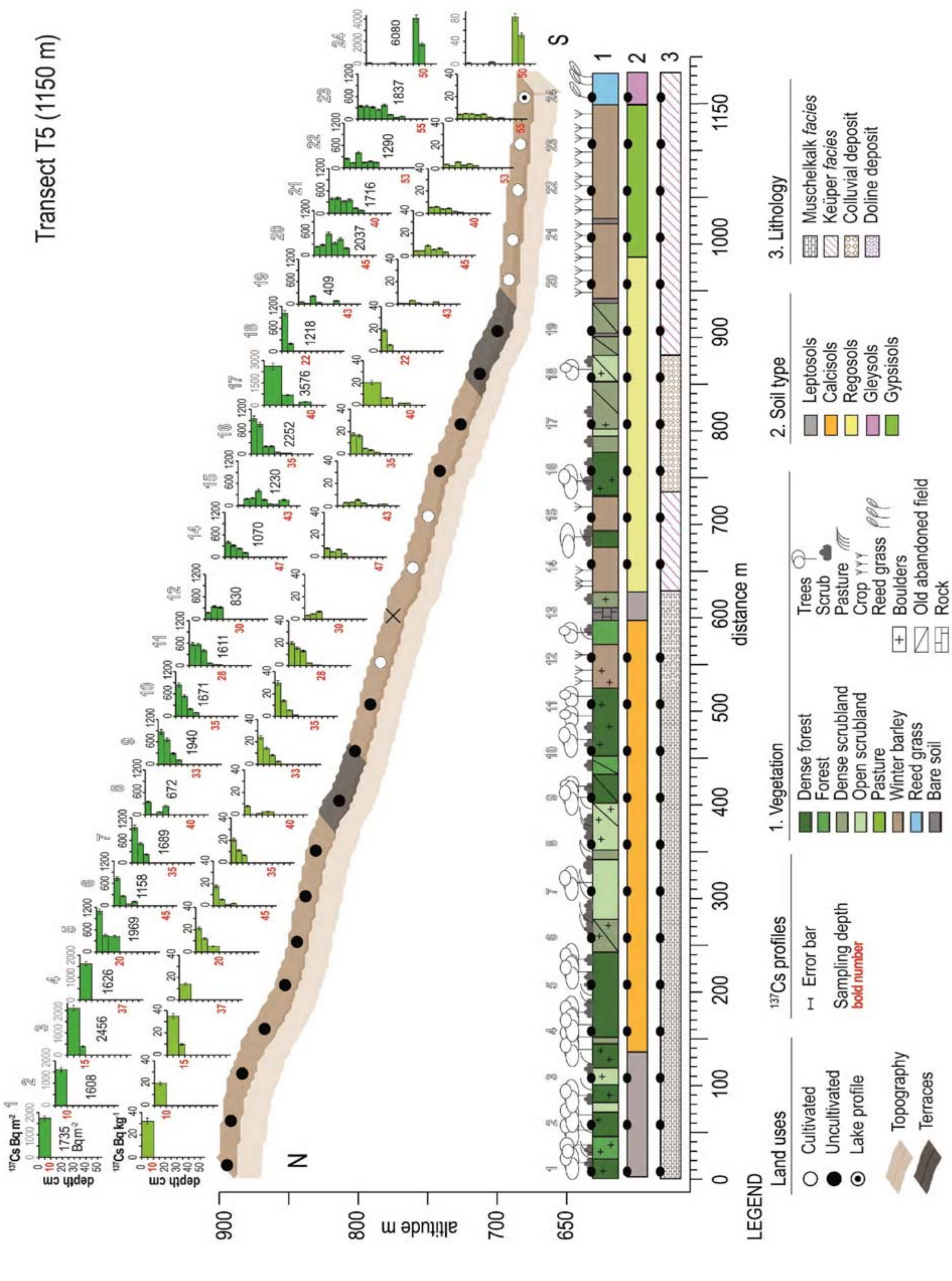

Figure 3. Depth distribution of ^{137}Cs mass activities and inventories for uncultivated and cultivated soil profiles.

"The central box covers the middle half of the data (extending from the lower quartile to the upper quartile); the whiskers show the location of the smallest and largest data values; the circles indicate outlier; the line within the box indicates the median of the data (50th percentile); and the plus sign (+) indicate the sample mean".

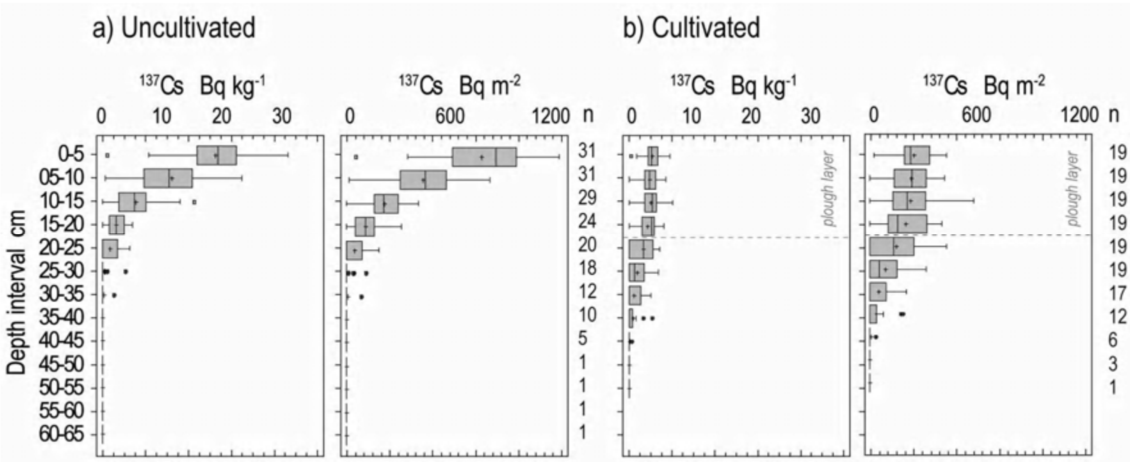

Figure 4. Depth distribution of physicochemical soil properties for uncultivated and cultivated soil profiles.

"The central box covers the middle half of the data (extending from the lower quartile to the upper quartile); the whiskers show the location of the smallest and largest data values; the circles indicate outlier; the line within the box indicates the median of the data (50th percentile); and the plus sign (+) indicate the sample mean".

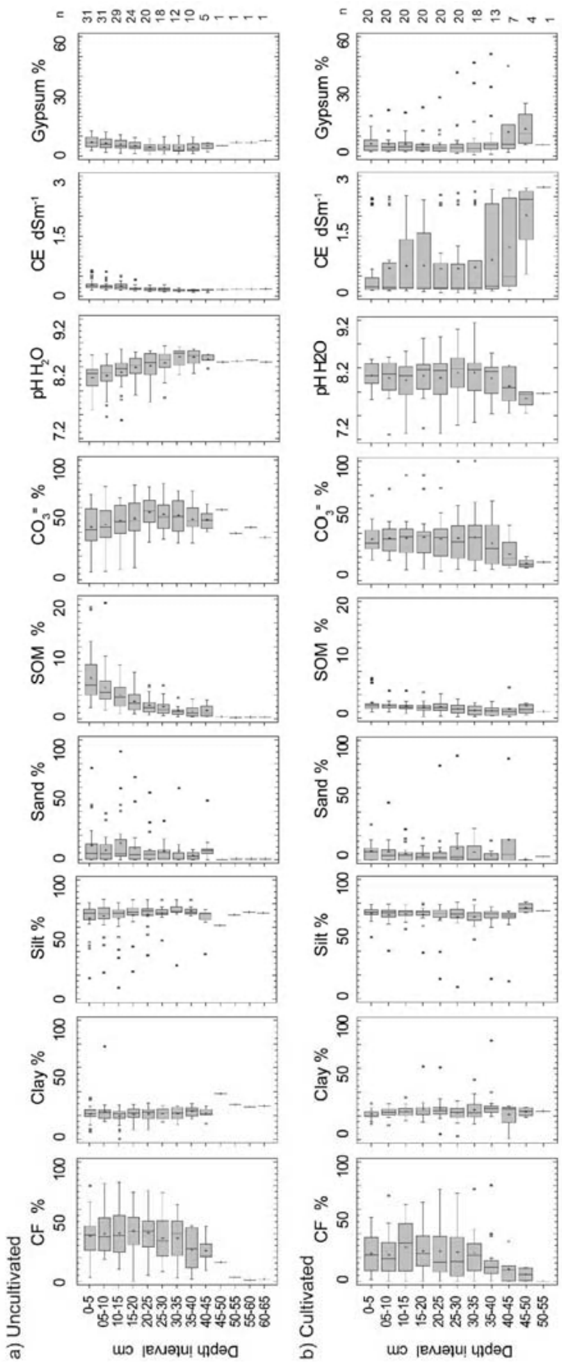

Figure 5. Boxplots of ^{137}Cs mass activities and inventories according to vegetation, soil type, lithology, altitude, slope gradient and orientation in uncultivated and cultivated soils. Different letters at the top of the box indicate significant differences at the p-level < 0.05 between different classes for each factor.

"The central box covers the middle half of the data (extending from the lower quartile to the upper quartile); the whiskers show the location of the smallest and largest data values; the circles indicate outlier; the line within the box indicates the median of the data (50th percentile); and the plus sign (+) indicate the sample mean".

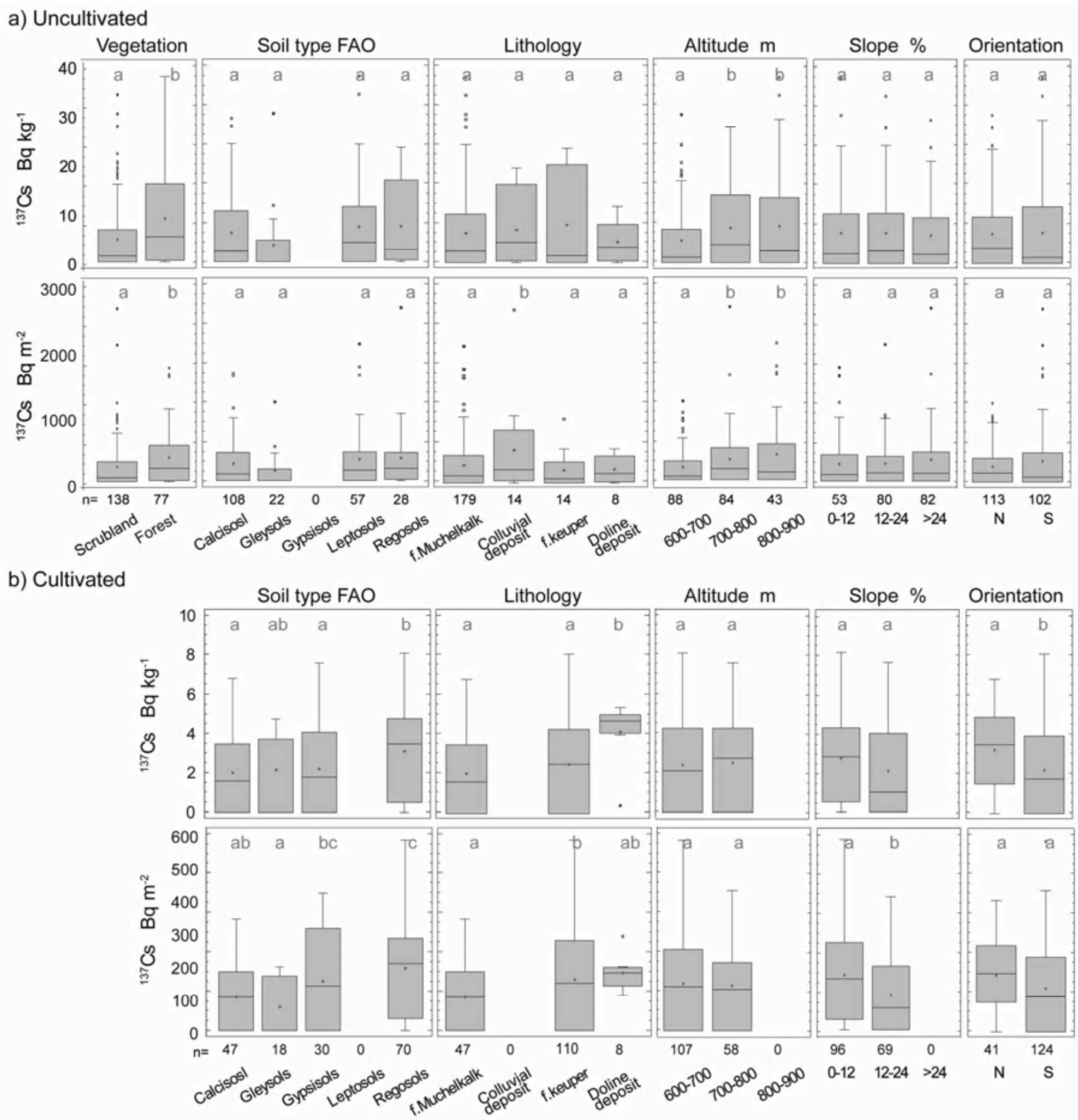

Figure 6. Dispersion diagram of soil interval samples after PCA analysis and principal components loadings, PC loading 1 vs. PC loading 2 for all interval samples.

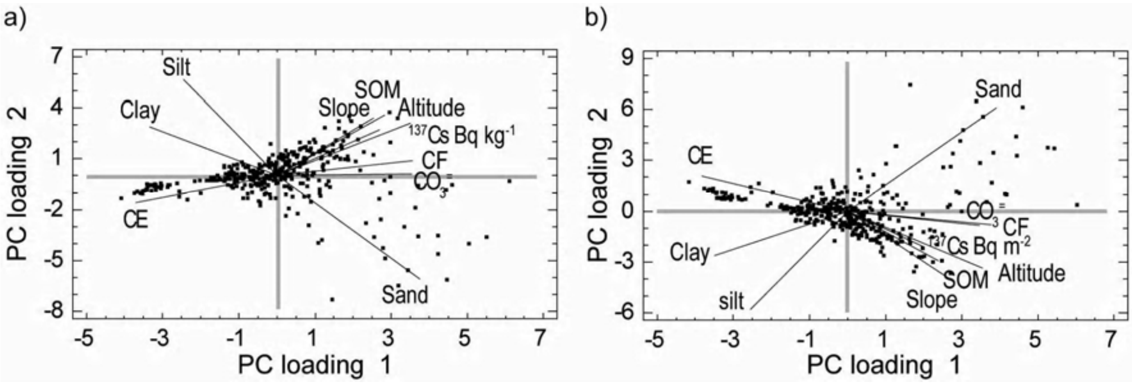

Figure 7. Observed (ordinate axis) and predicted values (abscissa axis) of ^{137}Cs mass activities and inventories for soil profiles based on a) equation 2 and b) equation 3.

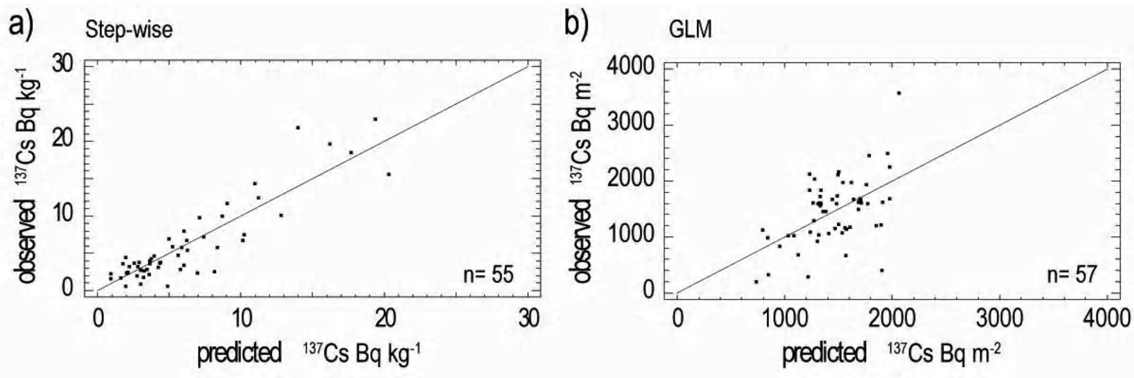

Figure 8. Observed (ordinate axis) and predicted values (abscissa axis) of ^{137}Cs mass activities and inventories for interval samples based on a) equation 5, b) equation 6 and c) equation 7.