**Spatial patterns of SOC, SON, ^{137}Cs and soil properties as affected by
redistribution processes in a Mediterranean cultivated field (Central Ebro Basin)**

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Abstract

Mediterranean mountain agroecosystems are sensitive areas to soil degradation due to climatic conditions and anthropogenic pressure from agriculture and grazing practices. Soil redistribution processes affect the spatial variability of soil properties and nutrients, as soil organic carbon and nitrogen (SOC and SON) are linked to soil quality and soil functions. A representative cereal field of rain-fed agroecosystems in a Mediterranean mountain environment where conventional and conservation agricultural practices have been implemented was selected on the northern border of the Ebro valley (NE Spain) to gain a better understanding of the effect of soil redistribution processes on the spatial pattern of soil properties at field scale. One hundred and fifty-six bulk (30–50 cm) and 156 topsoil samples (5 cm) were collected on a 10 x 10 m grid. Furthermore, 21 soil samples (40 cm) were collected at a nearby undisturbed reference site. ^{137}Cs massic activity was measured on bulk and reference soil samples, and SOC and SON were measured on all soil samples. The characterisation of SOC pools into active (ACF) and stable carbon fraction (SCF) was carried out to study SOC pool dynamics. The fallout ^{137}Cs was used in order to identify areas of ^{137}Cs loss and gain and that of soil within the study field. Similar spatial patterns and direct and significant correlations ($p \leq 0.01$) were found between ^{137}Cs and soil nutrients. As much as 70% of the total study field had lower values of ^{137}Cs inventory than the reference for the area. In addition, there was a generalised loss of soil nutrients. SOC gain was found in less than 1% of the study field and there was a large loss of SON compared to the undisturbed reference site. Higher and significant ($p \leq 0.01$) contents of soil nutrients were found in topsoil than in bulk samples. Furthermore, there was an enrichment of the relative contribution of ACF to total SOC in sampling points where there was a ^{137}Cs gain in both bulk and topsoil samples as a consequence of the soil redistribution particles by tillage and runoff.

Understanding patterns of soil nutrients can be useful for developing or implementing land management strategies to preserve soil quality in Mediterranean agricultural areas.

Keywords: Soil organic carbon; soil organic nitrogen; carbon pools; ^{137}Cs and soil redistribution; cultivated soils.

1. Introduction

The vulnerability of Mediterranean agroecosystems to soil erosion has been attributed to climatic regime characterised by irregular and scarce precipitation and drought periods, low vegetation cover and land-use changes (Bryan and Campbell, 1986). Soil erosion can be related to the loss of soil nutrients in the topsoil resulting in soil degradation, which, in turn, leads to reduced soil productivity and increased soil erodibility (Gregorich et al., 1998), and can be a serious threat to agricultural sustainability (Fornes et al., 2005). The severity of erosion processes in Mediterranean soils can be exacerbated by soil degradation through the loss of plant cover, biomass turnover and less soil organic carbon storage (Romanyà et al., 2006). Soil nutrients, such as soil organic carbon (SOC) and nitrogen (SON), represent one of the most important indicators of soil quality (Gregorich et al., 1994; Feng et al., 2007) and have an important influence on soil physical, chemical and biological properties (Monaco et al., 2008).

The production of SOC within soils occurs as a result of the addition of dead plant and animal material, and its loss occurs mainly due to the decomposition and mineralisation of soil organic matter (Polyakov and Lal, 2004). According to its chemical stability and turnover times, SOC can be divided into active carbon fraction (ACF), which has turnover rates of days to few years and is rapidly and easily decomposable (Coleman et al., 1996), and a more stable carbon fraction (SCF), with turnover rates ranging from a

few years to centuries and which is highly resistant to microbial and chemical decomposition (Falloon and Smith, 2000). The oxidation of the active carbon pool determines the soil-atmosphere exchange carbon fluxes; thus, this fraction influences the functioning of the ecosystems and nutrient cycling for maintaining the quality of soil and its productivity (Zou et al., 2005; Mazumdar et al., 2008). The stable carbon pool is responsible for long-term carbon storage in soils, which contributes to SOC storage. The characterisation of SOC fractions facilitates the understanding of the SOC pool dynamics, which is required to examine the ability of cultivated soils to stabilise carbon and its implications for soil degradation, especially in agricultural mountain Mediterranean landscapes that have been intensively cultivated for several centuries (Valero-Garcés et al., 2006).

SON is one of the most important nutrients for plants, and affects significantly plant growth, soil fertility and crop yield (Brady and Weil, 2002). The losses of soil nitrogen are due to the harvesting of crops, leaching, volatilisation, soil erosion and runoff (Schulten and Schnitzer, 1998; Udawatta et al., 2006). Further, SON plays an important role in carbon dynamics and is closely linked to the carbon cycle through biotic processes associated with vegetative productivity and organic matter decomposition (Post and Mann, 1990). The carbon/nitrogen ratio (C/N) is a good indicator of the degree of decomposition and quality of the organic matter held in the soil (Batjes, 1996).

Caesium-137 (^{137}Cs) is an environmental tracer that has been established as an effective and reliable tool to estimate soil redistribution in Mediterranean landscapes (Navas and Walling, 1992; Mabit et al., 2008; Benmansour et al., 2013; Navas et al., 2014; Porto et al., 2014). The redistribution of ^{137}Cs in the soil and across the land surface occurs in association with soil and sediment particles, and is controlled by its interaction with

land use practices, erosion and sediment transport processes (Walling and He, 1999). Tillage mixes the surface soil layers; therefore, ^{137}Cs and soil properties are uniformly distributed within the plow layer.

The relationships between the spatial variability of soil redistribution, soil properties and the SOC pool dynamics are still poorly understood and scarcely documented in Mediterranean mountain agroecosystems. Previous studies to relate soil movement traced with ^{137}Cs with the lateral variations of SOC and SON contents carried out in abandoned and cultivated soil profiles (Navas et al., 2012) and along a representative soil toposequence of Mediterranean mountain agroecosystems (Quijano et al., 2014a) encouraged further research to assess the spatial distribution of soil nutrients together with that of soil properties and ^{137}Cs at the field scale.

In order to address soil nutrient dynamics and soil movement closely related to the status of soil degradation, a crop field representative of Mediterranean agricultural landscapes was selected to examine the effects of soil redistribution processes on the spatial variability of soil nutrients and their relationship with topographic characteristics and soil properties. The specific objectives of this study were to: i) establish the reference inventories of ^{137}Cs and soil nutrients in soils of undisturbed areas under representative undisturbed Mediterranean vegetation cover, ii) to compare the reference inventories with the values of ^{137}Cs and soil nutrient inventories at the sampling points and establish the areas where loss and gain of ^{137}Cs fallout and soil nutrients has occurred within the field, iii) to characterise and measure active and stable carbon fraction contents to analyze the SOC pool dynamics in bulk and topsoil samples, and iv) to compare soil nutrient contents in the bulk and topsoil samples.

The knowledge of spatial patterns of soil nutrients and ^{137}Cs , and their relationships at the field scale are essential for understanding the impact of soil redistribution processes on soil nutrients and soil properties in Mediterranean mountain agroecosystems.

2. Materials and methods

2.1. Study site

The study site was a rain-fed crop field of 1.6 ha located in the central part of the Ebro Basin in the north-east of Spain (Fig. 1a) where winter cereals (*Triticum aestivum* and *Hordeum vulgare* L.) have been cultivated for the last 150 years under conventional tillage using a moldboard plow. The ploughing depth was between 25 and 30 cm in the sampling area; however, for the last fifteen years, reduced or minimum tillage (Stinner and House, 1990) was implemented and it was characterised by using a chisel reducing the number of tillage operations and leaving the crop residues on the surface as mulch. Liquid nitrogen fertiliser (18%) was applied in spring and autumn when precipitations are concentrated. The nitrogen fertiliser rates were 200 kg N ha⁻¹ for each application. The efficiency of N fertilisers depends greatly on the timing and intensity of rainfall in relation to the cropping period and on the timing of fertiliser application (Garrido-Lestache et al., 2004). In addition, splitting nitrogen fertiliser application between spring and autumn has been suggested as a strategy to improve wheat nitrogen use efficiency (López-Bellido and López-Bellido, 2001).

The topography of the study field was defined based on a detailed digital elevation model (DEM) together with field observations. The DEM with a 2.5 m resolution was created using a Total Topographic Station to obtain an elevation dataset on a regular 5×5 m grid (Fig. 1b). The average altitude of the study area was 630 m a.s.l, ranging from 622 to 636 m. The mean slope was 7.4%, ranging from 1.1 to 19%.

The climate is continental Mediterranean, characterised by summer drought and cold winters, and the rainfall events are frequent in spring (April and May) and autumn (September and October). The mean annual rainfall is around 500 mm and the mean annual temperature is 13.4°C. The study field was delimited by manmade infrastructures to the north (a paved trail and stonewall) and to the west (a drainage ditch), and it borders an ephemeral stream to the east and south. It comprises a hydrological unit (Quijano et al., 2013) that mostly remains isolated except from the occurrence of high storm events when it drains into the ephemeral stream (López-Vicente et al., 2015). The riparian system has a typical Mediterranean bottomland vegetation cover comprised of *Quercus coccifera*, *Quercus ilex* and *Populus alba*. Soils were classified as Calcisols (Quijano et al., 2014b), which are developed on Quaternary deposits mainly formed by alluvial deposits.

A field survey was done to select a representative site that complied with the characteristics of a stable reference area for assessing ^{137}Cs and soil nutrients under original vegetation cover and establishing the local reference inventories of ^{137}Cs , SOC and SON. The selected reference site was flat and undisturbed, it had not been affected by erosion or deposition processes and it was located near the study field to ensure similar ^{137}Cs fallout (Fig. 1c). Soils in the reference area were shallow and poorly developed Calcisols under typical Mediterranean vegetation cover with *Q. coccifera* and *Q. ilex* and scrubland (*Rosmarinus officinalis*, *Thymus vulgaris*, *Lygeum spartum* and *Santolina chamaecyparissus*).

2.2. Sample collection

At the reference site, 21 soil samples were collected until the parent material was reached, with depths ranging from 25 cm up to 40 cm. Sampling was done using a 7 cm

diameter automatic core driller and five samples were sectioned at 5 cm increments in order to study the vertical distribution of ^{137}Cs , SOC and SON.

In the cultivated field, 156 soil samples were collected using a 7 cm diameter automatic core driller on a 10 x 10 m grid (Fig. 1d). At each sampling point, one soil sample was collected at a depth of 30 cm, which corresponded to the plow layer. This sampling depth was considered to capture the effects of tillage and, consequently, soil homogenisation. The sampling depth was extended up to 50 cm in order to retain the entire ^{137}Cs profile in the sampling points identified as depositional sites according to field observations. The 30 and 50 cm soil samples are named, from now on, bulk samples. Furthermore, 156 topsoil samples were collected at the same sampling points of the bulk samples. The topsoil samples were taken with a 4 cm diameter manual core driller at 5 cm depth.

2.3. Laboratory analysis

Soil samples were air dried and weighed. Then samples were passed through a 2-mm sieve to separate the coarse fraction (>2 mm) and fine fraction (<2 mm) then they were packed, weighed, labelled and stored at 4°C prior to analyses. Analyses of physicochemical soil properties such as texture, pH, electrical conductivity (EC), carbonates (CO_3^{2-}), ^{137}Cs massic activity, SOC and its carbon fraction contents, active (ACF) and stable carbon fractions (SCF), and SON were performed on reference, bulk and topsoil fine fractions.

Particle size analysis was carried out with a Beckman Coulter LS 13 320 laser diffraction particle size analyser. Particle size distribution was determined after removing the organic matter by pre-treating the soil samples with H_2O_2 (10%) heated to 80 °C. The soil samples were then air dried and chemically disaggregated with 2 ml of

sodium hexametaphosphate (40%) and stirred for 2 h after that they were dispersed with ultrasound for a few minutes. The pH and EC (dS m^{-1}) were measured on saturated paste extracts of soil-water ratio, 1:2.5 and 1:5, respectively using a pH-meter and conductivity meter, respectively. Total carbonate content (%) was analysed using a calcimeter (CSIC, 1976).

SOC and SON contents (%) were measured with the dry combustion method using a LECO, *RC-612* multiphase carbon analyser and a LECO CN TruSpec carbon and nitrogen analyser, respectively. The dry combustion method is based on thermal oxidation of the sample at an established temperature. LECO devices consist of a furnace where a sub-sample of $<2\text{mm}$ soil (0.1500 g) grounded with a mortar and pestle is introduced and combusted into flow oxygen.

SOC content (%) was measured at $550\text{ }^{\circ}\text{C}$. The CO_2 gas evolved after combustion was determined by infrared analysis (Bremner, 1996; Nelson and Sommers, 1996). The decomposition of the most thermally labile components of SOC is released at approximately $300 - 350\text{ }^{\circ}\text{C}$ whereas decomposition of more refractory and stable carbon occurs at higher temperatures ($420 - 550\text{ }^{\circ}\text{C}$) (López-Capel et al., 2008). The characterisation of active carbon and stable carbon fractions was carried out using LECO, *RC-612* multiphase carbon analyser (Quijano et al., 2014a). The temperature of the furnace was stepped at $350\text{ }^{\circ}\text{C}$ and $550\text{ }^{\circ}\text{C}$ to oxidize the active (ACF) and stable carbon fractions (SCF), respectively. SON was measured by determining the NO_x gas evolved after combustion at $950\text{ }^{\circ}\text{C}$ by a thermal conductivity detector (LECO, 2006).

The SOC, SOC fractions and SON contents were converted into contents per surface area expressed as inventories or stocks (kg m^{-2}) multiplying the content by the mass of the fine fraction and dividing by the surface of the core sampler.

The ^{137}Cs technique is based on fallout of this anthropogenic radionuclide with a half-life of 30.2 years. The radionuclide was introduced into the stratosphere as a result of thermonuclear weapons test. The fallout of ^{137}Cs on land surface began in 1952 and continued to the mid-1970s, with a maximum peak in 1963, corresponding to the year of the Nuclear Test Ban Treaty, or nuclear accidents as Chernobyl in 1986 and Fukushima in 2011. The Iberian Peninsula was not significantly affected by the release from these nuclear accidents (García-León et al., 1993; Behrens et al., 2012). The technique assumes that the ^{137}Cs fallout is spatially uniform and its redistribution throughout the landscape is firstly controlled by its natural decay and by soil redistribution processes reflecting soil sediment transport (Walling and Quine, 1992). Once ^{137}Cs reaches the soil surface through wet and dry deposition is rapidly and strongly adsorbed by soil particles especially clay and soil organic matter (Tamura, 1964).

The massic activity of ^{137}Cs was measured using a high resolution, low background, low energy, hyperpure coaxial gamma-ray detector coupled to an amplifier and multichannel analyser. The detector had an efficiency of 50%, and a 1.9 keV resolution (shielded to reduce background), and was calibrated using standard soil samples in the same geometry as the measured samples. Gamma emission of ^{137}Cs was detected at 661.6 keV photopeak with counting times over 30000 s and the analytical precision of the measurements is $\pm 5 \%$ (95 % level of confidence).

The content of ^{137}Cs in the soil sample is expressed as a concentration or massic activity (Bq kg^{-1}) and as activity per unit area or inventory (Bq m^{-2}). The inventory was calculated using the mass of the fine fraction and the cross section of the core sampler (Navas et al., 2005).

2.4. Data analysis

The total inventory of ^{137}Cs for an individual sampling point was compared with the local reference inventory. Soil samples where inventories were lower than the local reference inventory were indicative of loss of radionuclides and, thus, soil loss. Similarly, inventories in excess of the reference level are indicative of the addition of radionuclides with soil particles by deposition (Walling and Quine, 1990). Similarly, the reference inventories of SOC and SON were compared with the SOC and SON inventories in the sampling points to determine whether a loss or gain of soil nutrients occurred in the sampling points.

Data were analyzed using SPSS 19.0 (Chicago, IL, USA). Principal component analysis (PCA) was used to discern relationships and groupings among the soil properties. PCA provides a useful means to analyze variance and reduce the dimensionality of a dataset consisting of a large number of interrelated variables, while retaining as much of the variation present in the data as possible. The number of components was determined using the eigenvalue-one criterion, which determines that components with eigenvalues greater than one and comprising variables with high component loading are to be extracted. The applicability of the PCA in the datasets used in this study was verified by applying Bartlett's sphericity and Kaiser-Meyer-Olkin (KMO) tests.

Pearson's correlation coefficients and linear regression analyses were used to assess the relationships between SOC, SOC fractions, SON, ^{137}Cs , topographic factors and soil properties. Furthermore, to assess the statistical significance of the effects of soil redistribution (eroded and depositional) and depth (bulk and topsoil) on soil properties at the sampling points, a one-way analysis of variance (ANOVA) was performed. Tukey's mean difference was used at the 5% probability level to separate the effects.

The ordinary kriging with trend was selected to model the spatial distribution of the measured soil physicochemical properties and SOC, SON and ^{137}Cs inventories at the field scale. All output maps and interpolations were performed using ESRI ArcGIS 10.2.1 software.

3. Results

3.1. Characteristics of the reference soils

Soils in the undisturbed area were shallow and poorly developed, and were derived mainly from Tertiary sandstones. A higher content of the coarse fraction from the parent material was found in soil layers at depths between 25 and 40 cm (Fig. 2).

Soils were alkaline (7.50 ± 0.14) and non-saline with an EC maximum of 0.44 dS m^{-1} and a mean value of $0.31 \pm 0.07 \text{ dS m}^{-1}$. Most soil samples had a silt loam texture, the silt and sand fractions were predominant; the mean values of silt and sand were $46 \pm 3.9\%$ and $42 \pm 4.7\%$, respectively. The clay fraction was less abundant with a mean value of $12 \pm 1.5\%$. Soils had a high coarse fraction content (mean = $13 \pm 3.7\%$) and carbonate content with maximum and mean values of 38% and $28 \pm 5.1\%$, respectively. The mean SOC content and inventory at the reference site were 1.6% and 5.4 kg m^{-2} , respectively and SON content and inventory were 0.34% and 1.4 kg m^{-2} , respectively. The reference ^{137}Cs inventory for the study area was $1507 \pm 92 \text{ Bq m}^{-2}$.

The reference profiles showed an exponential decrease of ^{137}Cs , SOC and SOC fraction contents and inventories from the surface to deeper soil layers (Fig. 2). ^{137}Cs massic activity was undetectable below 25 cm, whereas there was a high increase in the coarse fraction in deeper layers, mainly below 25 cm. The SON, clay, silt and sand contents were distributed relatively uniformly with depth and had no significant differences between the incremental depth intervals.

3.2. Soil properties in the study field

In the study field, soils were alkaline, non-saline and calcareous (Table 1). Most soil samples (81%) had a silt loam texture with a predominance of silt ranging between 50 and 71.9%. Thirteen per cent of the soil samples had loam and 3% had sandy loam textures with a relatively higher content of the sand fraction (35–74.3%). The remaining soil samples (3%) in which the clay content ranged between 36 and 56.5% had clay, silty clay and silt clay loam textures. The content of the coarse fraction varied from 0 to 17%. SOC contents were less than 1.8%, and the maximum SON content was 0.19%. In the field C/N ratios varied largely, ranging from 2 to 15 with a mean value of seven (Table 2).

The sand and carbonates increased from the upper (north-west) to the lower slope positions (south-east), whereas the opposite trend was found for clay, silt and EC (Fig. 3). The spatial pattern of pH was not as clear because the pH values had a low range of variation with no significant differences within the field. Fig. 4 shows the spatial distribution of SOC and SON contents (%). Higher SOC and SON contents were found on the upper slope, where higher clay values were also found. However, lower soil nutrient contents were found in the lower part of the field.

The PCA indicated that combinations of variables explained a relatively high proportion of the total variation between the samples. Results from 156 soil samples showed that 75% of the cumulative variance was explained by the first four components with eigenvalues greater than one (Fig. 5). The first component accounted for 30% of the total variance, corresponding to the positive and high values for SOC fractions, SON, ^{137}Cs and clay. The first component reflects the close relationship between soil nutrients, ^{137}Cs and clay. The second component, explaining 22% of the total variance,

was associated with the coarse fraction, sand and carbonates. The third component accounted for 14% of the total variance, corresponding to high positive loading values of EC and high negative loading values of pH, while the fourth component, which explained 9% of the total variance, identified high loading values of pH and SON (Table 3).

Most correlation analyses of the soil properties showed moderate to strong correlations (Table 4). ^{137}Cs , SOC, SOC fractions and SON inventories were directly and significantly correlated ($p < 0.01$). SOC and its carbon fractions were highly and directly correlated ($p < 0.01$) with the percentage of clay and silt, and highly and inversely correlated with the percentage of coarse fraction, sand and carbonates. On the other hand, the correlation between the coarse fraction, sand and carbonates was direct and statistically significant ($p < 0.01$).

3.3. The effect of soil redistribution processes on soil nutrients

In the study field, the mean value of the ^{137}Cs inventory was 1374 Bq m^{-2} (Table 2). Most soil samples ($n=98$) had lower values than the ^{137}Cs reference inventory and were identified as eroded points. Fifty soil samples had values higher than the reference inventory and were identified as depositional. Only eight soil samples had ^{137}Cs values in the range of the ^{137}Cs reference inventory for the study area. As expected, the means of ^{137}Cs massic activity and inventories in eroded points were lower and significantly different ($p \leq 0.01$) than in the depositional ones (Fig. 6). In addition, there was a significant difference ($p \leq 0.01$) between the mean values of the ^{137}Cs inventories in the sampling points collected at 30 cm ($1250 \pm 455 \text{ Bq m}^{-2}$) and at 50 cm ($1991 \pm 1107 \text{ Bq m}^{-2}$). Of the sampling points, 63% were identified as eroded, indicating a predominance of loss of ^{137}Cs , which affected 70% of the total area of the study field (Fig. 7).

The mean values of SOC and SON inventories were 3.5 and 0.51 kg m⁻², respectively (Table 2). Most soil samples had lower values of SOC than the reference inventory. Only 3% of the soil samples had higher values than the SOC reference inventory, which corresponded to less than 1% of the study field (Fig. 7). Moreover, a generalised loss of SON was found. Compared to the reference soils, the maximum value of SON inventory in the field (1.02 kg m⁻²) was lower than the SON reference inventory (Table 2). As much as 87% of the study field had a percentage of SON loss between 50 and 75% (Fig. 7). Examination of the spatial distribution of ¹³⁷Cs and soil nutrient inventories showed that similar patterns were found for ¹³⁷Cs and SOC. Despite the spatial pattern of SON being less coincident, the maximum and minimum values of SON inventories were found at the same sampling points of ¹³⁷Cs and SOC (Fig. 7).

When comparing eroded and depositional points, the means of clay and silt contents were lower at eroded points than at depositional ones, whereas the mean values of sand and carbonates were higher at depositional points, although there was not a statistical difference between them. However, the mean of the coarse fraction in the depositional points was higher and significantly different ($p \leq 0.05$) than in the eroded ones (Table 5).

The relationships between the percentages of loss and gain of ¹³⁷Cs and SOC were direct and significant ($r = 0.404$, $p \leq 0.01$) indicating that, for increasing percentage of ¹³⁷Cs loss, higher loss of SOC occurred.

On the other hand, soil samples at the eroded points presented significantly lower mean values of SOC, SOC fractions and SON inventories than at the depositional ones (Fig. 6). Furthermore, the relative contribution of ACF to SOC in the depositional points was found to be higher and significantly different ($p \leq 0.05$) than in the eroded ones. On the contrary, the relative contribution of SCF to SOC in depositional points was lower and significantly different ($p \leq 0.05$) than in the eroded ones (Fig. 6).

3.4. Soil nutrients in topsoil and bulk samples

Topsoil samples were characterised by higher and significantly different ($p \leq 0.05$) mean values of coarse and sand fractions than bulk samples. On the contrary, the mean values of clay and silt fractions were lower and statistically different ($p \leq 0.05$) in topsoil than in bulk soil samples. Moreover, there was not a significant difference between the mean values of carbonate contents in topsoil and in bulk samples (Tables 1 and 6).

Soil nutrients were more concentrated in the topsoil. The mean values of SOC, ACF, SCF and SON contents were higher and significantly different ($p \leq 0.05$) in topsoil than in the bulk samples; consequently, the C/N ratio was higher and significantly different ($p \leq 0.01$) in topsoil (Tables 2 and 6). Furthermore, the mean relative contributions of ACF/SOC and ACF/SCF in topsoil were higher and significantly different ($p \leq 0.05$) than in bulk samples. On the other hand, the relative contribution of SCF to SOC was higher and significantly different in bulk samples than in topsoil.

When comparing topsoil by soil redistribution processes, results were similar to that found for bulk samples. The means of clay and silt contents were slightly lower at eroded points than at depositional ones, whereas the mean values of coarse fraction and sand were higher at depositional points. However, the mean of carbonates in depositional points was slightly lower than in eroded ones, although there was not a statistical difference between them (Table 7). On the other hand, when SOC, SOC fractions and SON contents were compared in eroded and depositional topsoils, it was found that the mean values of SOC and ACF were higher and significantly different in depositional topsoil samples than in the eroded ones (Table 7).

4. Discussion

The depth distribution of ^{137}Cs in the reference profiles followed the typical pattern of undisturbed areas, as Gaspar and Navas (2013) also found in a nearby mountain agroecosystem. Most of the radiocaesium (>80%) was concentrated in the top 10 cm of the soil profile and declined rapidly with increasing depth reflecting the adsorption of the ^{137}Cs fallout by the surface soil and the limited post-fallout redistribution within the profile (Gaspar and Navas, 2014; Porto et al., 2003; Walling et al., 1995). The ^{137}Cs reference inventory for the study area falls within the range estimated by Caro et al. (2013) for the same area (1475 – 2288 Bq m⁻²) that obtained a ^{137}Cs deposition map for Peninsular Spain. According to Navas et al. (2007), the spatial distribution of ^{137}Cs showed a clear gradient with rainfall in the Central Ebro Basin. Such dependence of ^{137}Cs and rainfall is confirmed in the study area where the ^{137}Cs reference inventories compare to other areas with similar rainfall values (Navas et al., 2012, 2014) and are lower than ^{137}Cs inventories under higher rainfall, as in the axial Pyrenees (Navas et al., 2005).

The general exponential decrease of SOC and SOC fractions similar to that of ^{137}Cs in the reference profiles confirms that soils remain undisturbed under the original vegetation cover. The decrease of SOC with depth is likely associated with the processes of litter decomposition and carbon immobilisation which is more active in surface soil layers (Jobbágy and Jackson 2001).

In the study field, tillage mixes soil within the plow layer 0–25/30 cm and, in such way, cultivation produces the homogeneous distribution of ^{137}Cs and soil nutrients. In soil samples where there was a gain of ^{137}Cs , its depth distribution extended over 30 cm as a consequence of the accumulation of soil, and ^{137}Cs is found below the plow layer until a maximum depth of 50 cm.

The predominance of silt loam textures, which are highly erodible with low resistance to particle detachment and subsequent transport (Torri et al., 1997), together with low SOC contents (less than 2% in the study field), favours soil degradation and the depletion of soil quality as discussed by Loveland and Webb (2003). Therefore, these soils are prone to soil erosion by water and tillage, as indicated by a generalised loss of ^{137}Cs and soil nutrients compared to the inventories at the reference site.

Losses of SOC are less important than losses of SON. Seventeen per cent of the study field was affected by SOC loss ranging between 50 and 82%. Despite additions of nitrogen fertiliser in spring and autumn, 89% of its total surface had SON loss ranging between 50 and 81%. Once the fertiliser was applied, about 50% of the nitrogen was absorbed by crops (Craswell and Godwin, 1984). Apart from SON absorption and mineralisation (Schepers and Fox, 1989), the most important processes related to the loss of SON in the study field are leaching, volatilisation and export through runoff and soil erosion. The reason for lower SOC losses than SON could be the protective effect of cover mulching by reduced tillage practices carried out in the field over the last 15 years. Moreover, by adding nitrogen fertiliser higher crop biomass production could be related to an increase of SOC (Gregorich et al., 1996).

The important depletion of ^{137}Cs , SOC and SON is clearly associated with land use changes from forest to cultivated land. In addition, tillage practices and soil erosion by runoff are linked to the decline of soil nutrients affecting the soil structure and increasing soil susceptibility to erosive processes. The coincident spatial distribution of ^{137}Cs and soil nutrients suggested that similar redistribution processes affect their spatial patterns within the field. A main factor for soil redistribution in the study field is runoff through the gully system that drains the field into the main stream when exceptional rainfall events occur (López-Vicente et al., 2015). Runoff intensifies downslope,

triggering the selective removal of finer soil particles. Our results show that, at the lower end of the gully in the north-east part of the study field (Fig. 3), coarser and heavier particles are deposited as evidenced by an enrichment of the carbonate coarse fraction and sand in this part. The carbonate nature of the coarse fraction is confirmed by the direct and significant relationship between the coarse fractions, sand and carbonates. Eventually, the finer soil particles are exported out of the field to the main ephemeral stream by runoff.

Primarily, the soil's physical movement at the field scale affects the spatial distribution of soil particles within the field and, therefore, soil nutrients. Runoff redistributes preferentially the topsoil finer soil particles, like clay, which is associated with soil nutrients (Bajracharya et al., 2000), affecting their redistribution in the landscape (Lal, 2003; Zhang et al., 2011). In the study field, SOC and SOC fractions are associated with clay and silt size particles, which is confirmed by the direct and significant relationship between them, as also found by Zhao et al. (2006) for SOC in long-term cultivated soils. Thus, lower values of ^{137}Cs , SOC and SON appeared to be related to higher coarse fraction and sand contents at the lower end of the gully. According to Soto and Navas (2004, 2008), stoniness is one of the main features of soil in mountain Mediterranean agroecosystems and has to be taken into account for estimating the effective retention of ^{137}Cs . Furthermore, at the lower end of the gully, high percentages of ^{137}Cs and SOC loss are linked to the lowest contents of clay, silt and EC values.

The lack of significant correlations between ^{137}Cs and the finer fractions are due to the homogenous depth distribution of clay and the limited range of its variation in the study field, which is in agreement with the results found by Gaspar and Navas (2013) in a pre-Pyrenean catchment. The mobility of ^{137}Cs in temperate and in carbon organic poor soils, as in the study field, is primarily controlled by sorption onto clay mineral surfaces

(Sawhney, 1970). This highly selective sorption of ^{137}Cs occurs at the broken edges of the illitic-type clay minerals (Gaspar et al., 2014). However, organic matter whose major component is SOC is also important in the adsorption of ^{137}Cs , but this adsorption is supposed to be non-specific compared with the specific adsorption of ^{137}Cs in clays (Rigol et al., 2002). In agreement with Lee and Lee (2000), our results indicated that ^{137}Cs and SOC are closely related, which is supported by the direct relationship between ^{137}Cs , SOC and SOC fractions.

Agricultural practices for more than one hundred years have created a 20–30 cm deep furrow along the south-east border of the field, especially after the generalised use of heavy machinery since 1960. As a result of the plow furrow and soil compaction, the deposition of soil particles occurred in this part of the study field (Fig. 7). This fact was evidenced by the ^{137}Cs gain compared to the ^{137}Cs reference inventory and lower losses of soil nutrients with relatively high SOC inventories (4 and 4.5 kg m⁻²), and SON inventories between 0.6 and 1.02 kg m⁻². Navas et al. (2013, 2014) also found that the highest deposition occurred at the edges of cultivated fields.

The effect of soil redistribution processes on SOC dynamics is evidenced by the differences between the relative contributions of ACF and SCF to SOC in eroded and depositional points. The higher contributions of ACF to SOC in bulk and topsoil samples at depositional points suggested that, primarily, the smaller and lighter particles of soil enriched in the most bioreactive carbon fraction are removed preferentially downslope by physical processes related to water erosion. Furthermore, at eroded points, the soil loss led to the exposure of relatively enriched subsoil in the stable carbon fraction. Beguería et al. (2015) also found, in the central Ebro valley, a relevant role of the splash and wash off in the dynamics of SOC in agricultural soils with an

enhancement of enriched SOC soil particles in the splashed material with respect to the original soil under natural precipitation.

Analyses of the behavior of SOC and SOC fractions in relation to the loss and gain of ^{137}Cs and, therefore, of soil were not conclusive. However, the trends of the relationships between the loss ratio of $^{137}\text{Cs}/\text{SOC}$ and the relative contributions of ACF and SCF to SOC ($r = -0.160$, $r = 0.161$, respectively) could suggest that a relatively higher loss of ^{137}Cs versus SOC would be associated with a decrease in the contribution of ACF to SOC and an increase of the ratio SCF/SOC.

When considering points where soil accumulation was high, which was done by selecting sampling points with more than 60% ^{137}Cs gain ($n=13$) in relation to the ^{137}Cs reference inventory, the relationships between the ^{137}Cs inventory and the ratios of ACF/SOC and SCF/SOC suggested that, for increasing ^{137}Cs , the relative contribution of SCF to SOC increases ($r=0.392$), while the opposite occurred for ACF/SOC ($r=-0.391$). The protection of soil organic carbon by stabilisation of microaggregates could be an important process at depositional sites. The prevalence of more stable carbon fractions at depositional sites is related to the buried SOC leading to a higher degree of SOC mineralisation as described by Boix-Fayos et al. (2015) across several deposit compartments in Mediterranean catchments (SE Spain).

However to confirm these trends, further analyses of the behavior of SOC and SOC fractions in relation to a loss and gain of ^{137}Cs , and that of soil with a larger dataset are required.

The enrichment of soil nutrients in topsoils compared to bulk soil samples was in agreement with results by Quijano et al. (2014a) in cultivated soil profiles studied along a mountain toposequence representative of Mediterranean agroecosystems. The incorporation of crop residues into soils by conservation tillage practices implemented

in the study field favors the enhancement of soil nutrients restricted to the soil surface (0–5 cm depth), as also reported by Blanco-Moure et al. (2013) and López et al. (2012) in the central Ebro valley. Despite the migration of finer soil particles into underlying layers as a consequence of cultivation (Yonkeu et al., 2000) and the exportation of finer soil particles by wash off, the accumulation of soil nutrients in topsoils is recorded in the study field. This fact reinforced that the implementation of reduced tillage is a good measure to increase SOC in topsoils. Besides, this SOC increase will influence other factors, such as infiltration, water content, porosity and soil aggregation (Veum et al., 2011), improving soil quality (Govaerts et al., 2009). In addition, higher ACF in depositional topsoil samples than in eroded ones suggested that, in agreement with Carter (1992), the more bioreactive carbon fraction was more sensitive to soil management than was stable carbon.

5. Conclusions

In the study field, the spatial patterns of ^{137}Cs and soil nutrients are affected by similar soil redistribution processes closely related to agricultural management practices and runoff. The generalised loss of ^{137}Cs and soil nutrients relative to the values of the reference inventories is mainly due to the exportation of finer soil particles enriched in ^{137}Cs and nutrients out of the field through the gully system. The selective removal of finer soil particles mobilised the rich bioreactive carbon fraction, and this was associated with a higher contribution of ACF to SOC at depositional sites for both topsoil and bulk soil samples. From the point of view of soil quality and soil organic carbon sequestration, the incorporation of crop residues into soil by reduced tillage practices was a good measure for increasing SOC in topsoils. The present study may help to improve the current scarce knowledge of the relationships between the spatial

variability of soil redistribution and soil nutrient patterns and dynamics in Mediterranean agroecosystems, which requires further research to mitigate soil degradation and implement erosion control practices at the field scale.

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Table 1 Basic statistics of the physicochemical soil properties in the bulk samples of the study field.

n=156	>2 mm (%)	clay (%)	silt (%)	sand (%)	pH	EC (dS m ⁻¹)	CO ₃ ²⁻ (%)
Mean	1.09	19.00	57.59	23.42	8.25	0.17	37.90
Median	0.44	18.85	60.25	20.45	8.25	0.16	38.15
S.D.	2.19	5.98	9.98	13.31	0.13	0.04	3.12
CV %	201.22	31.48	17.33	56.84	1.62	26.01	8.24
Min	0.00	5.30	18.90	3.50	7.90	0.07	31.35
Max	16.85	56.50	71.90	74.30	8.67	0.46	48.82

S.D.: standard deviation; CV: coefficient of variation

Table 2 Basic statistics of ^{137}Cs and soil nutrients in the bulk soil samples of the study field

n=156	^{137}Cs (Bq kg ⁻¹)	^{137}Cs (Bq m ⁻²)	SOC (%)	SOC (kg m ⁻²)	ACF (%)	ACF (kg m ⁻²)	SCF (%)	SCF (kg m ⁻²)	ACF/SOC	SCF/SOC	ACF/SCF	SON (%)	SON (kg m ⁻²)	C/N
Mean	2.45	1373.72	0.64	3.49	0.42	2.30	0.22	1.18	0.66	0.34	2.05	0.09	0.51	7.13
Median	2.43	1278.84	0.64	3.43	0.42	2.27	0.21	1.14	0.66	0.34	1.95	0.09	0.47	7.05
S.D	0.96	668.17	0.19	0.97	0.13	0.63	0.07	0.38	0.04	0.04	0.65	0.02	0.16	2.03
CV %	39.01	48.64	30.49	27.82	30.31	27.21	33.82	31.84	6.54	12.88	31.52	23.83	31.01	28.49
Min	n.d.	0.00	0.13	0.96	0.12	0.85	0.02	0.11	0.51	0.11	1.03	0.05	0.26	2.35
Max	5.72	4092.09	1.83	7.34	1.20	4.81	0.63	2.54	0.89	0.49	7.85	0.19	1.02	14.63

S.D.: standard deviation; CV: coefficient of variation; n.d.: not detected

Table 3 Principal component loading (PC_i) for the factors with eigenvalues. >1.

	PC ₁	PC ₂	PC ₃	PC ₄
>2 mm %	-0.30	0.35	0.02	0.02
¹³⁷ Cs Bq kg ⁻¹	0.24	0.39	-0.33	-0.42
¹³⁷ Cs Bq m ⁻²	0.25	0.50	-0.23	-0.15
clay %	0.30	-0.35	-0.25	-0.16
sand %	-0.34	0.39	0.19	0.16
ACF kg m ⁻²	0.45	0.18	0.08	0.21
SCF kg m ⁻²	0.50	0.03	0.10	0.24
CO ₃ ²⁻ %	-0.21	0.30	-0.19	0.05
SON kg m ⁻²	0.28	0.26	0.28	0.47
pH	-0.14	-0.12	-0.41	0.61
EC dS m ⁻¹	0.02	-0.01	0.66	-0.24

Table 4 Pearson correlation coefficients between soil properties of the bulk samples.

n=156	> 2 mm (%)	clay (%)	silt (%)	sand (%)	CO ₃ ²⁻ (%)	¹³⁷ Cs (Bq kg ⁻¹)	¹³⁷ Cs (Bq m ⁻²)	SOC (kg m ⁻²)	ACF (kg m ⁻²)	SCF (kg m ⁻²)
clay %	-0.396									
silt %	-0.590	0.350								
sand %	0.621	-0.712	-0.907							
CO ₃ ²⁻ %	0.397	-0.255	-0.303	0.342						
¹³⁷ Cs Bq kg ⁻¹	0.004	0.035	0.033	-0.040	0.084					
¹³⁷ Cs Bq m ⁻²	0.088	-0.107	-0.073	0.103	<u>0.158</u>	0.846				
SOC kg m ⁻²	-0.275	0.310	0.267	-0.340	<u>-0.186</u>	0.327	0.411			
ACF kg m ⁻²	-0.216	0.255	0.200	-0.265	-0.084	0.358	0.439	0.980		
SCF kg m ⁻²	-0.351	0.375	0.356	-0.435	-0.339	0.246	0.329	0.944	0.860	
SON kg m ⁻²	-0.074	-0.070	0.019	0.017	-0.047	0.091	0.411	0.535	0.504	0.538

Bold numbers and underlined numbers indicate statistical significance at p<0.05 level. Bold numbers indicate statistical significance at p<0.01.

Table 5 Mean and standard deviation of the bulk soil properties by soil redistribution.

	eroded (n=98)	depositional (n=50)
>2 mm %	0.76±1.38a	1.76±3.24b
Clay	19.35±5.89a	18.02±5.35a
Silt	58.67±9.58a	55.77±10.86a
Sand	21.98±12.59a	26.21±14.95a
CO ₃ ²⁻ %	37.45±3.12a	38.52±3.15a

Different letters indicate significant differences at p<0.05 level

Table 6 Basic statistics of the soil properties for the topsoil samples.

n=156	>2 mm (%)	Clay (%)	Silt (%)	Sand (%)	CO ₃ ²⁻ (%)	SOC (%)	ACF (%)	SCF (%)	ACF/SOC	SCF/SOC	ACF/SCF	SON (%)	C/N
Mean	2.91	11.62	52.28	36.09	38.12	1.02	0.71	0.30	0.70	0.30	2.41	0.12	9.86
Median	1.42	11.80	53.15	35.05	38.18	1.00	0.71	0.30	0.70	0.30	2.37	0.12	8.59
S.D.	5.16	2.00	5.63	7.31	2.67	0.26	0.18	0.09	0.03	0.03	0.54	0.05	9.49
CV %	177.23	17.17	10.76	20.24	7.00	25.72	25.16	28.93	4.27	10.05	22.59	39.19	96.29
Min	0.00	5.10	25.70	24.60	28.98	0.29	0.25	0.03	0.64	0.11	1.79	0.01	2.53
Max	38.20	15.30	61.60	69.20	46.05	1.88	1.27	0.61	0.89	0.36	8.08	0.37	92.11

S.D.: standard deviation; CV: coefficient of variation

Table 7. Mean and standard deviation of the topsoil properties by soil redistribution.

	Eroded (n=98)	Depositional (n=50)
>2 mm %	2.34±3.28a	4.13±7.74a
Clay %	11.79±1.99a	11.38±2.08a
Silt %	52.64±5.49a	51.64±6.24a
Sand %	35.56±7.16b	36.98±8.04b
CO ₃ ²⁻ %	38.13±2.48a	37.99±3.05a
SOC %	0.99±0.24a	1.08±0.30b
ACF %	0.69±0.16a	0.76±0.21b
SCF %	0.30±0.08a	0.32±0.10a
ACF/SOC	0.70±0.03a	0.71±0.04a
SCF/SOC	0.30±0.03a	0.29±0.04a
ACF/SCF	2.36±0.31a	2.49±0.85a
SON %	0.12±0.05a	0.13±0.05a
C/N	9.79±8.36a	10.24±12.08a

Different letters indicate significant differences at p<0.05 level

Fig. 1. a) Location of the study field in the central part of the Ebro basin (NE Spain), b) DEM of the study field with flow lines in the gully and ephemeral main stream, c) Photo of the reference site and the study field, d) 3D view of the sampling points on a 10 x 10 grid.

Fig. 2. Depth distribution of the ^{137}Cs , SOC, SOC fractions and SON, coarse fraction and soil texture in the reference site.

Fig. 3. Spatial distribution of the grain size fractions, pH, EC and carbonate content in the study field.

Fig. 4. Spatial distribution of SOC and SON contents in the study field.

Fig. 5. Dispersion diagram of bulk samples (n=156) after PCA analysis.

Fig. 6. Box-plots of ^{137}Cs massic activity and inventory, SOC, SOC fractions and SON contents and inventories in eroded and depositional sampling points.

Fig. 7. Spatial distribution of ^{137}Cs , SOC and SON inventories. Contour lines and plots show the loss and gain percentage of ^{137}Cs , SOC and SON inventories in the study field.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7