Effects of land use on soil organic and inorganic C and N at ^{137}Cs traced erosional and depositional sites in mountain agroecosystems

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Highlights

- Soil redistribution and land use are key factors on nutrients stock
- Highest SOC and TN stocks occur in cropland at depositional sites
- Labile carbon is more sensitive than stable fraction to soil erosion and land use
- Cropland lowlands are important sinks of soil inorganic carbon
- ^{137}Cs data help to understand soil nutrients pools in Mediterranean agroecosystems

Abstract

Patterns of erosion, transport and deposition of soil particles not only affect the distribution of mineral fraction of the soil but their organic components. The spatial variability of soil nutrients in Mediterranean mountain agroecosystems that combine complex land uses and abrupt topography is poorly documented despite emerging concerns on nutrients fate require understanding the potential impact of soil erosion on nutrients redistribution across landscapes. This study aims to assess at catchment scale the spatial patterns of soil organic and inorganic carbon (SOC, SIC), soil organic carbon fractions and total nitrogen (TN) in relation to land uses and soil redistribution. To this

purpose we selected a well characterized endorheic catchment located in NE Spain where soil erosion and deposition rates derived from ^{137}Cs measurements were previously documented and we mapped the distribution of cropland and rangeland areas. A grid of 100x100 m was established for collecting a total of 205 soil samples for nutrients and standard soil analyses.

In both rangeland and cropland the patterns of SOC and TN stocks were clearly related to ^{137}Cs inventories thus significantly lower nutrient stocks (SOC: $34.1 \pm 18.6\text{Mg ha}^{-1}$, TN: $2.9 \pm 1.7\text{Mg ha}^{-1}$) were found in erosional sites whereas higher stocks (SOC: $44.9 \pm 24.2\text{Mg ha}^{-1}$, TN: $3.9 \pm 2.3\text{Mg ha}^{-1}$) were recorded in depositional areas. In cropland the mean nutrients contents (SOC: $1.1 \pm 0.8\%$, TN: $0.1 \pm 0.1\%$) were significantly lower than in rangeland (SOC: $3.2 \pm 2.1\%$, TN: $0.2 \pm 0.1\%$). The highest C/N ratios (29) were found in rangeland, while in cropland they occurred in depositional areas with their values below 10 in the erosional sites. In turn, the SIC content (range 0.4 to 10.2%) was slightly lower at the depositional sites ($4.9 \pm 2.2\%$) but not significantly different from the erosional sites ($5.1 \pm 2.2\%$). Cultivated and steep slopes led to loss of soil and nutrients due to intense erosion processes also related to tillage and crop productivity. Our findings provide evidence that the combined effect of land use and soil redistribution exert an important control on the spatial distribution of nutrients, highlighting a significant decrease of SOC and TN stocks with increasing soil erosion in croplands. The information gained achieves a better understanding of soil nutrients dynamics and further knowledge for effective land management of mountain agroecosystems in the Mediterranean region.

Keywords: Nutrients; land uses; soil redistribution; ^{137}Cs ; spatial patterns; NE Spain

1. Introduction

The soils in the Mediterranean region of southern Europe have low values of organic matter ($\leq 2\%$), being very low ($\leq 1\%$) in approximately 75% of the whole surface, and in this way they are exposed to progressive degradation processes (Aranda and Comino, 2014). Soil organic matter, which is composed of around 58% of soil organic carbon (SOC), influences several ecosystem services such as soil formation, soil aggregation and structure and nutrient storage (Johnston et al., 2009). Increasing SOC content improves different functions of soils like soil structure, soil water holding capacity and soil fertility, and quantifying these functions requires prediction and an spatially explicit representation of the distribution of SOC (Adhikari et al., 2019). The SOC may be affected by different factors including land use, soil management, land degradation, climate, hydrology, soil type and Geomorphology (Hancock et al., 2010). Land use and hillslope characteristics play important roles in influencing soil water erosion which is the primary factor for the depletion or restoration of SOC stock and hence may cause CO₂ emissions or sequestration (Nosrati et al., 2015).

Soil management actions and strategies are aimed at carbon sequestration (Lal, 2009; Wells et al., 2019). For example conversion of cropland to pastoral and forest lands (Auerswald and Fiener, 2019), conventional tillage to conservation and no tillage (Balesdent et al., 2000), no manure use to regular addition of manure (Maillard et al., 2016), and to soil specific fertilisation rate, terracing, natural green covers adding organic matter or natural green covers (Chenu et al., 2019; Novara et al., 2019). Further, in recent years recommended management practices have been implemented to increase SOC stocks (Almagro et al., 2017). Since soil is the largest reservoir of C in terrestrial ecosystems, a slight variation in this pool can result in substantial changes in the atmospheric CO₂ concentration, thus potentially affecting global climate change

(Davidson and Janssens, 2006; Gao et al., 2017). The “4 per 1000” initiative which aims to reach a SOC stock annual growth of 4‰ with a view to altering that of total atmospheric carbon (Francaviglia et al., 2019; Minasny et al., 2017; Novara et al., 2017).

Knowledge on the spatial distribution of soil organic carbon for different land uses incorporating erosion on hillslopes is essential for estimating SOC distribution and stock. Moreover, controlling factors on carbon storage in soil profiles can be used to plan soil management strategies and changes in land use to mitigate carbon loss (Boix-Fayos et al., 2015). The ability of soil to store and sequester organic carbon depends to a great extent on the balance between OC input and losses from soil and by complex interactions of SOC pools (Lal et al., 2015). SOC pools can be arbitrarily separated into different fractions based on recalcitrance (Zimmermann et al., 2007). Quijano et al. (2016a) divided SOC into two distinct carbon fractions by the dry combustion method: the active and decomposable fraction (ACF) with rapid turnover rates, which acts as a short-term carbon reservoir, and the stable carbon fraction (SCF) with lower turnover rates that acts as a long-term reservoir. Numerous national and regional databases of soil properties and associated estimates of soil carbon stock consider the organic, but not the inorganic carbon (SIC) fraction (Rawlins et al., 2011). Research on spatial distribution of soil IC concentrations in calcareous soils is scarce. Many soils in Mediterranean regions of Europe are developed on calcareous parent material and contain large amounts of carbonates, i.e. inorganic carbon (SIC). Therefore, SIC must be taken into account in order to not overestimate the actual content of soil carbon and report any future change in soil carbon stock resulting from the formation of pedogenic carbonates (Marchant et al., 2015).

Soil erosion has significant impacts on the distribution and transformation of SOC within a landscape (e.g. Van Oost et al., 2007). Soil erosion preferentially removes SOC

(especially the more labile materials) from topsoils in upslope eroding positions and the deposition of carbon enriched sediments lead to accumulate SOC in downslope positions (Lal, 2003). The impact of soil erosion in the carbon budget and the implications with regard to soil management to maximize sequestration is under debate (Borrelli et al., 2018; Lugato et al., 2018; Berhe et al., 2007). There is still a controversy on the role of erosion areas acting as source of carbon/nutrients and depositional ones as being sinks. In the light of the controversy over the role of erosion in the carbon budget and the implications with regard to soil management to maximize sequestration, there is a need to examine the relationships between SOC distribution and soil redistribution (Zhang et al., 2006).

Worldwide land use changes have been identified as main factors causing soil erosion (e.g. Kachanosky, 1987) and decline in soil properties (Navas et al., 2008). This has been further confirmed in Mediterranean landscapes (Navas et al., 2017; Lizaga et al., 2019) and it has been proved that also produces loss of soil carbon (Boix-Fayos et al., 2009; Quijano et al., 2017).

Fallout Caesium-137 (^{137}Cs) is a well-established tracer of soil erosion in the Mediterranean region (Porto et al., 2011; Gaspar et al., 2013a; Navas et al., 2013; Lizaga et al., 2018) and it provides a valuable tool for investigating medium-term erosion-carbon relationships (Zhang et al., 2006). In soil profiles, fallout radionuclides (FRNs: ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$) show a depth distribution highly coincidental with that of SOC (Gaspar and Navas 2013; Gaspar et al., 2017) as organic matter is a main binding agent of these radionuclides in the soil fine components (Gaspar et al., 2013b). Based on ^{137}Cs and SOC profiles that are well described in Mediterranean soils (e.g. Quijano et al., 2016a; Mesrar et al., 2017) it can be inferred that soil erosion will also affect SOC and TN as main components of the organic matter. Furthermore, eroded particles that are

distributed in the landscapes by water and tillage processes show clear links between ^{137}Cs rates of soil erosion and losses of soil nutrients as it has been documented in agricultural fields (Ritchie and McCarty, 2008; Martinez et al., 2010; Navas et al., 2012; Quijano et al, 2016b). The relationships between spatial patterns of soil erosion and soil nutrients for small agricultural landscape units and cultivated fields by using fallout ^{137}Cs has been evidenced in a variety of environments (Wei et al., 2008; Teramage et al., 2013; Li et al., 2018). Besides, studies have been conducted on the pathways of ^{137}Cs and SOC transport in channel bed sediments (Boix-Fayos et al., 2017) but few have explored the spatial patterns of soil redistribution and nutrients variations at catchment scale.

The spatial variability of soil carbon including inorganic C stocks and nitrogen in relation to physiography and land use in sloping Mediterranean mountain area still remains poorly documented (Ramos and Martínez-Casasnovas, 2006) and more research is needed to improve the current findings. In this context, this study aims to assess the effect of land use and soil redistribution on nutrients dynamics in an endorheic karstic catchment located in the Marginal Pyrenean Range in NE Spain. To this end, we examine the spatial distribution of SOC, SOC pools (active and stable carbon fractions, ACF and SCF, respectively), SIC and TN stocks in rangeland and cropland areas by using the rates of soil loss/gain derived from ^{137}Cs measurements that were previously documented in the area. The comprehensive information gained from evaluating the relationships between soil erosion and soil nutrients will help to interpret the effect of land uses and soil redistribution behind the spatial distribution of nutrients which is key for improving our understanding on soil nutrients pools in Mediterranean mountain agroecosystems.

2. Material and Methods

2.1. Study area

The study area is located in the central part of the Spanish Pre-Pyrenees. The catchment is characterized by an endorheic karst system with several dolines induced by evaporite dissolutions that host two permanent lakes at the lower part of the catchment (Fig. 1). This area is known as Estaña lakes and was declared by European NATURA 2000 Network as a Site of Community Importance (SCI). The climate is typical Mediterranean continental with mean annual rainfall of ca. 460 mm concentrated in autumn and winter and mean annual temperature of 12.2 °C (Gaspar et al., 2013a).

The catchment (246 ha) is characterized by abrupt topography, with altitudes between 676 to 894 m and mean slope of 18%. Steep slopes (>20%) occupy 35% of the catchment while flat areas are in the low part surrounding the lakes. Numerous linear elements are distributed across the landscape. These are natural elements like scarps and vegetation strips but also human interventions like unpaved trails and stone walls for agricultural terraces.

A complex mosaic of land uses (Navas et al., 2014) dominated by Mediterranean open forest of *Quercus* and submediterranean plants, winter barley as the main crop, scrubland, pasture areas and riparian vegetation cover the landscape. Two main land uses have been classified and grouped for this research; rangeland (R) and cropland (C). Rangeland covers almost 60% of the catchment preferably located at the headwaters and steep slopes areas while cropland (C) occupies gentle slopes mostly surrounding the lakes taking 43% of the catchment (Fig.1).

The geological substrate of the study area is made of Mesozoic materials, outcrops of Keüper and Muschelkalk Facies. The soil types are described in detail in Machín et al. (2008) and Gaspar et al. (2014). Following a typical toposequence from the catchment

divides to the bottom lakes the main soil groups are represented by Leptosols (30 %), Regosols (23 %) and Calcisols (32 %) on the Muschelkalk Facies. Next patches of Gypsisols (9 %) developed on Keuper facies extend over the flat cropland areas and Gleysols (3%) are distributed surrounding the lakes (Fig.1).

2.2. Sample collection

A total of 205 soil cores were sampled across the catchment on a ca. 100 m grid. The depth of the cores varied from 15 to 55 cm in function of the soil type (Fig 1). According to land use, 141 soil samples were collected under rangeland (R) and another 64 under cropland (C). From fallout ^{137}Cs the soil redistribution rates obtained in previous studies (Navas et al., 2013, 2014) allowed to classify a total of 115 sites as erosional (E) where soil has been lost and another 90 sites that accumulated soil are classified as depositional (D). Combining both factors the soil samples were grouped into: C-E n=40, C-D n=24, R-E n=75, R-D n=66.

2.3 Laboratory analyses

Soil samples were air-dried, weighed and sieved to $\leq 2\text{-mm}$ and stored at 4°C prior to analysis of study soil parameters. The soil organic carbon (SOC) and the active (ACF) and stable (SCF) carbon fractions were analyzed by using LECO RC-612 multiphase carbon analyser (LECO Corporation, St. Joseph, MI, USA). The characterisation and fractionation SOC fractions (ACF and SCF) were carried out following protocols by Quijano et al. (2016a). Soil inorganic carbon (SIC) content (%) was analysed using the pressure-calimeter method (CSIC, 1976) and TN was measured by Kjeldhal method (Bremner, 1996).

The contents of SOC, ACF, SCF, SIC and TN were converted into stocks (Mg ha^{-1}) according to the following equation:

$$\text{Property Stock (Mg ha}^{-1}\text{)} = \text{Property content (\%)} * \text{BD} * \text{D} * 100$$

where BD is the soil bulk density (g cm^{-3}), property content (%), and D is the soil sampling thickness (m).

Particle size analysis was carried out with a Beckman Coulter LS 13 320 laser diffraction particle size analyser (Beckman Coulter, Inc., 2011), after removing the organic matter by pre-treating the soil samples with H_2O_2 (10%) in a boiling water bath at 80°C and adding 2 ml of solution of a dispersing agent (40%, sodium hexametaphosphate (NaPO_3)₆) to avoid grain flocculation (Murray, 2002). 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 (Hanna, Instruments, Italia Srl., PD) and gypsum content were determined following standard techniques (CSIC, 1976).

Measurements of ^{137}Cs mass activity were performed by using a high resolution, hyperpure germanium, coaxial gamma-ray detector coupled to an amplifier and multichannel analyser (Canberra Xtra, Canberra industries, Inc. USA) following the methodology described by Navas et al. (2005). Estimates of soil redistribution rates derived from ^{137}Cs inventories were obtained applying the conversion model reported by Soto and Navas (2004 a,b, 2008) for uncultivated and cultivated soils.

2.4. Statistics

Correlations and principal component analysis (PCA) were performed to analyse the relationships between nutrients and ^{137}Cs and main soil properties. Data were analyzed using SAS/STAT (9.1 SAS Inst. Inc., Cary, NC, USA) by SAS (1998). The

effects of the land uses (two levels; cropland C - rangeland R) and soil redistribution (two levels; erosion E - deposition D) on soil nutrient parameters were analyzed using the least square methodology of the General Linear models GLM procedure, with treatment (land use and soil redistribution factors) as the fixed effect. Interactions between fixed effects were tested before applying the final statistical model and were found to be no significant. Least square means are reported throughout using *sums of squares Type III*, and P values <0.05 were deemed to be statistically significant. Homogeneity of variance was tested before analysis, assuming normal distribution.

3. Results

3.1. Relationship between nutrients and soil properties

In the study catchment soils are alkaline and non-saline. The mean contents of SOC, ACF and SCF were 2.5%, 1.8% and 0.8%, respectively. SOC reached contents up to 15%, corresponding to ACF the highest contents while SCF did not exceed 2.6%. Mean TN content was 0.19% with a maximum below 1%. The SIC contents ranged from 3% to 6% in the 85 per cent of the soil samples with a mean content of 4.9%. Soils are stony in average amounts around 27%, reaching 69%. Most soil samples (85%) had a silt loam texture with a predominance of silt ranging between 52% and 80%, while the rest of samples had silty clay loam, loam, loamy sand and sandy loam textures. The mass activity and inventories of ^{137}Cs in the catchment soils ranged from 0.5 to 43.8 Bq kg^{-1} and 58 to 6398 Bq m^{-2} , respectively.

Significant differences were found between land uses for all nutrients, while differences between erosional and depositional sites were different only for SCF and TN (Fig. 2). The content of SOC, ACF, SCF and TN in soils gradually decreased from the rangeland depositional sites (R-D) to the rangeland erosional sites (R-E), followed

by cropland depositional sites (C-D) and cropland erosional sites (C-E) (Fig. 2). Stoniness and sand content presented similar behaviour as nutrients and the content of ^{137}Cs Bq kg⁻¹ also showed the same trend, even clearer than nutrients since differences for land use and soil redistribution, were highly significant. The mean SIC content did not differ between groups. On the contrary, pH was significantly lower in rangeland erosional and depositional sites than in cropland, where pH was slightly higher in erosional than depositional sites. Other properties like EC and gypsum were not significantly different despite mean values were slightly higher in erosional than in depositional sites (Fig. 2).

For each land use, Table 1 shows the correlations between nutrients and soil properties. In rangeland, SOC, SOC fractions and TN were directly and significantly correlated with ^{137}Cs activity ($p \leq 0.05$) and inversely correlated with pH ($p \leq 0.05$). On the contrary, SIC% was directly correlated with pH ($p \leq 0.05$) and presented an inverse correlation with ^{137}Cs activity, gypsum content and EC ($p \leq 0.05$). In addition, SOC, SCF and TN were directly and significantly correlated with silt content ($p \leq 0.05$) and inversely correlated with sand content. Similarly, in cropland, significant direct correlations were found between SOC, SOC fractions, SIC and TN with ^{137}Cs activity ($p \leq 0.05$). Similarly like in rangeland, SIC showed an inverse correlation with EC and gypsum. The main discrepancy between both land uses was the lack of correlation between nutrients and particle size fractions in cropland.

The PCA indicated that the same combinations of properties explained a relatively high proportion of the total variation for rangeland (75.4%) and cropland (80.1%) (Fig. 3). For both land uses, the cumulative variance was explained by the first three components with eigenvalues greater than one (Fig. 3). The first component reflected the close relationship between soil nutrients and ^{137}Cs and also with the finer fractions

(clay and silt) and negative loading values of sand. The second component was associated with the pH and SIC, while the third component corresponded to high positive loading values of EC and gypsum. The main difference between land uses was that cropland showed closer associations between silt and clay in first component and SIC and pH in the second component.

3.2. Soil redistribution in the land uses

The mean value of the ^{137}Cs inventory in the catchment was 1531 Bq m^{-2} . By comparing the local reference inventory of ^{137}Cs ($1570 \pm 80 \text{ Bq m}^{-2}$; Gaspar et al., 2013a) with the point ^{137}Cs inventories, 56% of the sampling points had values lower than the local reference inventory indicating loss of ^{137}Cs and, thus, soil loss. In 44% of the sites ^{137}Cs inventories exceeded the ^{137}Cs reference inventory indicating soil deposition.

The erosion and deposition rates were significantly higher in cropland than in rangeland soils. The soil redistribution rates ranged from a maximum value of erosion rate of $140.5 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ to a maximum deposition rate of $83.6 \text{ Mg ha}^{-1} \text{ yr}^{-1}$. More than half of sampling points in cropland (62%) and in rangeland (53%) experienced soil erosion with a mean value of $47.9 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ and $7.2 \text{ Mg ha}^{-1} \text{ yr}^{-1}$, respectively. Soil deposition was recorded in 38% and 47% of the sampling points of cropland and rangeland, respectively, with a mean value of $36.0 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ in cropland and $3.6 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ in rangeland (Table 2).

3.3. Factors controlling the distribution of nutrients stocks

The highest nutrient stocks were found in cropland depositional sites, while the lowest stocks were recorded in erosion sites. The mean value of SOC and SCF stocks

was significantly higher in cropland depositional sites than in the other groups, which presented similar stocks among them with almost half of SOC and SCF stocks in rangeland and cropland erosional sites (Table 3). Similarly, the mean stock of ACF was significantly higher in C-D than in R-D sites and this, in turn, was significantly higher than in R-E and C-E sites. The mean of TN stock was significantly different between each group, decreasing gradually from C-D to C-E, followed by R-D and R-E sites. Similarly to TN, the mean SIC stock was significantly higher in C-D than in C-E, but in turn they were significantly higher than in rangeland, in both erosional and depositional sites (Table 3).

The spatial pattern of nutrients stocks shows in general good agreement with that of ^{137}Cs inventories (Fig. 4). The SOC, ACF, SCF and TN stocks were highly variable. High SOC and TN stocks were found both at the upper part of the catchment where ^{137}Cs inventories are closer to the reference inventory as well as at the bottom part where croplands are located, especially at depositional sites. SIC stock was highest at the bottom part in croplands. The ^{137}Cs inventories decreased from hilltop to bottom showing that higher soil loss occurred at lower slope positions in coincidence with the presence of croplands. On the terraced fields, there were abrupt changes in ^{137}Cs inventories over very short distances. In rangeland soil redistribution rates were low to moderate. Croplands registered both the highest erosion and deposition rates. High soil erosion was found in both flat and steep areas, however high deposition rates were restricted to flat areas (Fig. 4).

The mean relative contributions of SOC fractions to the total SOC content (ACF/SOC and SCF/SOC) show that ACF/SOC did not differ between erosional and depositional sites in cropland whereas in rangeland the relative contribution of ACF to SOC in depositional sites was significantly higher than in erosional sites (Fig. 5). On the

other hand, in rangeland ACF/SOC was significantly higher ($p \leq 0.05$) than in cropland soils whereas SCF/SOC was significantly lower in rangeland than in cropland. The C/N ratios varied largely, ranging from 3 to 29 (mean: 12), but since SOC and TN were higher in rangeland soils consequently, the C/N ratio was higher and significantly different ($p \leq 0.05$) in rangeland than in cropland soils.

4. Discussion

The strong effect of land use change on nutrient depletion supported by the fact that the contents of SOC, ACF, SCF and TN are almost three times higher in rangeland than in cropland sites have significant implications to nutrient export, vegetation cover and nutrient management linked to restore agricultural degraded soils in Mediterranean landscapes. Other studies have also found important decreases of nutrients after transformation of natural forests to croplands (FAO, 2016, Lizaga et al., 2019). Soil disturbance, depletion of nutrients and high erosion rates ($48 \text{ Mg ha}^{-1} \text{ y}^{-1}$) associated to the long history of agricultural practices show the intense degradation of these Mediterranean soils. Cultivation and tillage practices associated to the depletion in C/N ratio are crucial for the growth and functioning of crops with adverse impacts on ecosystem services (Pennock et al., 1994; Hancock et al., 2010). In addition, the lower C/N ratio (<10) found in erosional sites of agricultural soils is a main factor which affects the availability of SOC and agricultural sustainability (Melero et al., 2009; Sun et al., 2018) thus the intensive use of Mediterranean mountain calcareous soils over many decades promotes low resilience of these soils in terms of SOC recovery as also found by Romanyà and Rovira (2011).

Similar processes affect the spatial distribution of ^{137}Cs and soil nutrients (SOC and TN) as shown by the coincidence between their distribution patterns and the correlation

of ^{137}Cs , soil redistribution and SOC content underlining the relation between these parameters also reported in several studies (Mabit et al., 2008; Navas et al., 2012; Gaspar and Navas, 2013; Quijano et al., 2016b). Furthermore, soil TN is closely coupled to SOC evidencing the similar pathways of these organically bound nutrients through the soil in cropland and rangeland soils (Table 1).

On the other hand, the highest mean nutrient stocks (SOC and TN) in cropland depositional sites is determinant to characterize these areas as a sink of soil nutrients, besides the physiographic characteristics of the catchment which in turn controls soil redistribution leading to soil accumulation at specific parts of lowlands and hillslopes. Soil redistribution processes appear closely linked to topography affecting the variability of the soil redistribution rates and soil nutrient distribution along the slope. In agreement with Li et al. (2018) and Navas et al. (2017) the slope gradient was an important parameter affecting soil erosion and SOC distribution. Moreover, in the Estaña catchment, there is large variability of soil redistribution rates because sudden changes in ^{137}Cs inventories appear in areas of terraced fields. The abrupt shifts in ^{137}Cs inventories are accompanied with similar changes in nutrient stocks related to the presence of landscape linear elements but reflecting also the effect of surface curvature. Thus high nutrients and ^{137}Cs contents found in steep slope areas occur in depressional spots before stone walls on terraced fields. Other studies like Zhang et al. (2006) observed that boundaries across the slope seem to create lines of zero downslope transport of soil resulting in accumulation of soil and SOC on the upslope side of the boundary.

The main reason for the higher TN stock in cropland compared to rangeland is attributed to fertilisation. Once fertiliser is applied, about 50% of the nitrogen is absorbed by crops (Craswell and Godwin, 1984). The most important processes related

to the loss of TN apart from absorption and mineralisation (Schepers and Fox, 1989), are leaching, volatilisation and export by runoff (Quijano et al., 2016a). In our catchment soil water erosion should be also considered an important cause of TN loss in step croplands. The lowest TN stock in rangeland may be attributed to N loss through leaching which karstic processes could explain. Besides Leptosols where most of the rangeland sampling points is located, are thin soils and their good drainage conditions may further facilitate leaching thus counterbalancing the inputs of natural vegetation and resulting in relatively poor TN content.

The higher SIC stocks found in cropland compared to rangeland could be linked to the accumulation of inorganic carbon made available from eroding sites at the headwaters where calcareous soils are abundant, along with dissolved inorganic carbon mobilized through the soil profile by surface and subsurface water (Quijano et al., in press). In addition, cropland extends over Calcisols on argillaceous Keuper materials where the SIC pool consists of primary inorganic carbonates and secondary inorganic carbonates. Therefore, besides land use the reason of lowlands for being a SIC sink has an important pedogenetic origin further promoted by the existing alkaline environment as also reported by Wang et al. (2015). Other studies have also shown that SIC content is largely related to land use type, thus SIC stock was significantly higher in cropland than in grassland in the Yunwu Mountain, China (Liu et al., 2014) and in Russian Chernozems (Mikhailova et al., 2016). To date, however, there are few studies assessing the relationships between SIC and SOC, which led to inconsistent findings. In that respect, different climate conditions and variations in soil properties which are related to transformation and accumulation of SIC and SOC may explain the strong positive correlation between SIC and SOC as reported by Guo et al., 2016.

In general, we find high erosion rates on relatively gentle slopes with predominance of cropland soils. Besides, most cultivated fields are on Calcisols with low mean SOC content. However, according to previous studies (Gaspar and Navas, 2013; Quijano et al., 2016a) most rangeland soils are Leptosols located upslope that have the highest SOC content and low soil erosion rates. In the catchment different soil types are present because of significant differences in Geomorphology, vegetation, parent materials, and cultivation patterns (Gaspar et al., 2014). Furthermore, we have to consider the natural evolution of the soil catena from the upper part of the slopes to the bottom part with increasing soil thickness. Starting from the headwaters with predominant thin, well drained Leptosols followed by Regosols the sequence ends with better developed and thicker Calcisols with patches of Gypsisols. Thus most croplands that are at the bottom part have thicker soils also in coincidence with the argillaceous parent material and the accumulation of soil from eroded upper slopes with implications on soil and nutrients distribution in the landscape. These soils are in turn closely associated with land use and, therefore, affect the lateral distribution of SOC along the slopes which is also in agreement with the results by Dengiz et al. (2015) who identified the primary factors influencing the spatial distribution of SOC at catchment scale. In our agricultural fields, water and tillage erosion are two main processes of soil redistribution as found in several environments (Van Oost et al., 2006; Quinton et al., 2010; Quijano et al., 2016c). In other Mediterranean agroecosystems (Nadeu et al., 2015; Nosrati et al., 2015) soil erosion driven by land use seems also to be determinant for the SOC redistribution (Chappell et al., 2015).

The biogeochemical cycles of carbon fractions are strongly influenced by land use since it affects their storage and distribution. In cropland soils, the relative contribution of ACF and SCF to SOC is lower than in rangeland because soil disturbance and mixing

during tillage practices linked to the disruption of soil aggregates increase the availability of the most active fraction of SOC to microorganisms which may result in a decrease of ACF and in a release of carbon dioxide (CO₂) back to the atmosphere (Olson et al., 2017). On the other hand, in rangeland soils, it is remarkable the significant and higher relative contribution of ACF to SOC in depositional sites than in erosional sites, where SCF/SOC was significantly higher. Soil changes after soil erosion processes may affect substantially the most active fraction that is preferentially lost when soils are eroded leading to a relative enrichment in SCF to SOC. Because ACF consists of the mineral-free soil organic matter which turns over rapidly, is responsible for much of the biological activity in soil (Six et al., 2002). ACF is easily affected by land use management as well as erosion therefore the labile carbon is a more sensitive indicator for evaluating changes in SOC than other SOC pools in agreement with the results from Sainepo et al. (2018). Further, in a southern Mediterranean catchment Boix-Fayos et al. (2009) also found that land use induced changes in SOC and SOC fractions which determine whether catchments behave as sources or sinks of soil carbon.

Land uses besides soil redistribution exert an important control of the spatial distribution of nutrients in this topographically driven catchment. Assessing the role of land use along with erosion and deposition on the nutrients distribution is critical not only for understanding future challenges posed by climate change but also for providing mitigation strategies.

5. Conclusions

The results of this study demonstrate variability in nutrients contents and stocks across the study catchment that shows different patterns in cropland and rangeland. Nutrient content of erosion sites was significantly lower than that of depositional sites.

The ¹³⁷Cs measurements proved very useful to help understand nutrient dynamics, which has been possible from sampling the entire soil profile. Our results demonstrated the importance of taking into account soil thickness for accurately assessing spatial variation of nutrients stocks.

The spatial pattern of nutrients stocks was strongly affected by the physiography of the catchment that is a key factor of soil redistribution (erosion or deposition). The finding that the highest erosion and deposition rates occur in croplands instead of rangeland and that they are associated to lowest and highest nutrients stocks, respectively, point out to land use besides soil redistribution as key controls of the spatial distribution and availability of soil nutrients.

Our results reflect that soil redistribution and land use had biogeochemical implications in SOC dynamics. Labile carbon is more sensitive to soil redistribution and land use than stable carbon fraction. Rangeland soils had higher relative contributions of ACF and SCF to SOC than croplands. The decomposable labile C fraction was preferentially removed at erosional sites and accumulated in depositional sites where there was significantly highest relative contributions of ACF to SOC. Cropland lowlands are important sink of SIC related with its pedogenetic origin in this alkaline environment.

Identifying the driving factors that influence the spatial distribution of soil nutrients is important to better understand their patterns at the catchment scale. This knowledge is especially relevant for soil monitoring and modelling nutrients response in the landscape. The insight gained about soil nutrients patterns in erosional and depositional sites is essential for improving our knowledge about sink and sources of soil nutrients of interest to improve effective soil management of Mediterranean mountain landscapes.

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Figures

Figure 1. Location of the study area and the soil sampling points collected with the land use, soil type and topography maps of the study catchment.

Figure 2. Boxplot of the nutrients content, main soil properties, ^{137}Cs measurements and redistribution rates for cropland and rangeland at erosional and depositional sites.

Figure 3. Dispersion diagram plots and principal components loadings of the three first components from a PCA for (a) cropland and (b) rangeland soil samples.

Figure 4. Spatial distribution of nutrients stocks and ^{137}Cs inventories within the catchment.

Figure 5. Boxplot of ACF/SOC, SCF/SOC and C/N for cropland and rangeland at erosional and depositional sites.

Table 1. Pearson correlation coefficients between each pair of nutrients content, main soil properties and ^{137}Cs measurements. The upper triangle shows the correlation values for the rangeland samples (n=141) and the lower triangle shows the correlation values for the cropland samples (n=64). Bold and shaded numbers indicate statistical significance at the 95.0% confidence level, *P-values* below 0.05.

	SOC %	ACF %	SCF %	TN %	SIC %	Clay %	Silt %	Sand %	^{137}Cs Bq kg ⁻¹	^{137}Cs Bq m ⁻²	pH	EC dS m ⁻¹	Gypsum %
SOC %		0.987	0.681	0.939	-0.157	-0.009	0.174	-0.166	0.583	0.168	-0.406	0.122	0.001
ACF %	0.995		0.556	0.925	-0.152	-0.028	0.141	-0.133	0.548	0.143	-0.411	0.140	0.023
SCF %	0.983	0.960		0.649	-0.118	0.083	0.258	-0.251	0.522	0.218	-0.232	-0.006	-0.099
TN %	0.916	0.895	0.932		-0.189	0.041	0.209	-0.209	0.593	0.230	-0.385	0.092	0.003
SIC %	0.224	0.237	0.194	0.126		-0.195	-0.063	0.145	-0.084	-0.070	0.194	-0.392	-0.538
Clay %	0.123	0.123	0.120	0.126	-0.175		0.620	-0.814	-0.027	-0.037	0.057	-0.026	-0.034
Silt %	0.038	0.013	0.085	0.086	-0.234	0.630		-0.902	0.026	-0.098	-0.133	0.106	-0.046
Sand %	-0.074	-0.056	-0.107	-0.110	0.234	-0.833	-0.955		-0.037	0.101	0.099	-0.072	0.045
^{137}Cs Bq kg ⁻¹	0.569	0.598	0.500	0.445	0.326	0.109	0.047	-0.075		0.668	-0.179	-0.122	-0.070
^{137}Cs Bq m ⁻²	0.152	0.157	0.139	0.176	0.132	0.104	0.210	-0.190	0.651		0.008	-0.214	-0.067
pH	-0.298	-0.305	-0.277	-0.248	0.460	-0.155	-0.182	0.189	-0.297	-0.142		-0.679	-0.358
EC dS m ⁻¹	-0.166	-0.161	-0.171	-0.156	-0.514	0.042	0.111	-0.095	-0.039	0.058	-0.671		0.628
Gypsum %	-0.116	-0.104	-0.136	-0.149	-0.484	0.217	0.075	-0.136	-0.060	-0.022	-0.465	0.589	

Table 2. ^{137}Cs inventory and erosion and deposition rates in terms of two different types of land use.

	<i>Cropland C</i>					<i>Rangeland R</i>				
	<i>n</i>	<i>mean</i>	$\pm SE$	min	max	<i>n</i>	<i>mean</i>	$\pm SE$	min	max
^{137}Cs Bq m ⁻²	64	1479.97	1099.78	92.08	6398.16	141	1554.46	727.12	58.14	3753.37
Erosion Mgha ⁻¹ y ⁻¹	40	47.92	42.91	2.10	140.54	75	7.17	11.40	0.10	65.10
Deposit Mgha ⁻¹ y ⁻¹	24	35.96	27.35	0.70	83.60	66	3.59	3.52	0.10	17.70

Table 3. Least square means of nutrients stocks (Mg ha ⁻¹) for land use and soil											
redistribution. Different letters in the same row indicate significant differences, p≤0.05.											
A: Land use			<i>Cropland C</i>				<i>Rangeland R</i>				
B: Soil Redistribution			<i>Erosion E</i>			<i>Deposition D</i>			<i>Erosion E</i>		
			C - E <i>n</i> = 40			C - D <i>n</i> = 24			R - E <i>n</i> = 75		
			<i>mean</i>	± <i>SE</i>		<i>mean</i>	± <i>SE</i>		<i>mean</i>	± <i>SE</i>	
SOC stock			33.40	3.27	a	58.09	4.22	b	34.51	2.38	a
ACF stock			21.13	2.34	a	36.94	3.01	c	23.77	1.71	ab
SCF stock			12.27	1.14	a	21.15	1.47	b	10.74	0.83	a
SON stock			3.93	0.27	c	6.06	0.35	d	2.47	0.20	a
SIC stock			190.96	10.67	b	233.16	13.78	c	67.58	7.79	a