Variability in ^{137}Cs inventories and potential climatic and lithological controls in the central Ebro valley, Spain.

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Abstract

The extent of soil erosion in some Spanish semiarid regions has caused great concern regarding the sustainability of soil resources. Accelerated soil erosion, particularly in some Mediterranean areas, is likely to be one of the main environmental problems associated with climate change. Fallout ^{137}Cs has been shown to provide a reliable basis for assessing soil erosion rates in different environments around the world. However, existing information concerning the spatial variation of ^{137}Cs inventories at reference sites has identified a need for further investigation of the factors affecting their spatial variability in semiarid environments, where stony and skeletal soils are predominant. Reference sites at three locations in the central Ebro valley were selected to investigate the ^{137}Cs content of

several grain size fractions. Each site included both natural vegetated conditions and cultivated land and the three sites were characterized by different values of mean annual rainfall. The results obtained demonstrate the influence of lithology, land use and climate on the spatial variability of ^{137}Cs inventories that increase from 1190, to 1500 and 1710 Bq m^{-2} with increasing annual rainfall values from 300 to 500 mm at the study sites. The soils on marls at the Valareña site had the highest proportion of ^{137}Cs in the coarse fractions of cultivated soils (12 %) in comparison with soils developed on limestones at Loma Negra (5 %), whereas no ^{137}Cs content was found in the coarse fractions of soils on glaciis-terrace materials at Peñaflo. The ^{137}Cs reference inventories are higher in soils on marls and sands at cultivated locations at Valareña and Peñaflo, but have similar values in soils at cultivated and uncultivated locations on limestones at Loma Negra. Therefore, in absence of level undisturbed soils with natural vegetation cover, cultivated flat soils on hard rocks could provide reliable reference inventories.

Key words: ^{137}Cs reference inventories, physiographic factors, lithology, rainfall, land use, semiarid regions, central Ebro basin, Spain.

Introduction

Some semiarid regions in Spain, such as the central part of the Ebro valley, are suffering from severe erosion. The extent of this problem has caused much concern about the future of the fragile soil resources, and has emphasized the urgent need to implement soil conservation measures. A first step in this direction is to quantify actual erosion rates. The application of the ^{137}Cs technique to investigate erosion rates and patterns, provides a

means of assessing the movement of soil particles within a globally-applicable worldwide framework. This radioisotope has been widely used in many different environments around the world ^{1,2,3}. From the first studies ^{4,5,6} to more recent investigations^{7,8}, the ¹³⁷Cs technique has been applied successfully to trace soil movement and other environmental processes operating over the last 30 years.

Fallout ¹³⁷Cs has proved to be a reliable method for quantifying erosion processes in highly fragile environments, such as the semiarid steppe and temperate mountain areas in the central part of the Ebro basin ^{9, 10}. Research carried out in the region¹¹ (Las Bardenas area) showed that erosion rates on cultivated land were five times higher than on uncultivated land. In these existing studies, analyses of the spatial distribution of the ¹³⁷Cs inventories highlighted the need for further investigation of the factors affecting the spatial variability of ¹³⁷Cs reference inventories in these semiarid environments⁹, where stony and skeletal soils with high gravel content are predominant. This issue has also been subject of research in Australia ¹².

The work reported in this paper investigates the ¹³⁷Cs levels found at three study sites located in the central Ebro valley. Each site includes both natural vegetated conditions and cultivated land under different mean annual rainfall. The aim is to analyze the influence of climatic, lithological and pedological factors on the local variability of ¹³⁷Cs reference inventories. Among the lithological and pedological factors the nature and size of soil particles and aggregates may influence the final estimation of ¹³⁷Cs inventories. In order to assess this effect ¹³⁷Cs measurements are performed in several grain size fractions. These data may provide the basis for a better understanding of ¹³⁷Cs behaviour in these semiarid environments that are characterized by irregular patterns of surface stoniness and vegetation cover, which affect the initial distribution of the radioisotope.

Study sites

Three sites located along a climatic transect and representative of the most typical physiographic conditions found in the central Ebro valley were selected for this study (Figure 1). The main physiographic and land use characteristics of the three study sites are summarised in Table 1.

The landscape of the central Ebro basin is characterized by high Tertiary plateaux dissected by a fluvial network, draining towards the River Ebro. The Loma Negra site, named after one of these central plateaux, lies at an altitude of 600 m above sea level. This structural platform has carbonate materials at the hilltop on which Rendzic Leptosols have developed. These soils are poorly developed and their depth does not exceed 20-25 cm. Due to the flat terrain, much of the Loma Negra plateau is under cultivation for cereals, but in some areas relicts of a natural vegetation of pine trees (*Pinus halepensis*), *Quercus coccifera* and other shrubs species still remains.

The Valareña site is located at the foot of the Loma Negra plateau at an altitude of 400 m above sea level. It is underlayed by Miocene sandstones, marls and clays. The soils according to the FAO (Food and Agriculture Organization of the United Nations) classification are Leptosols. This level area is also intensively cultivated for agricultural crops, and very little natural vegetation remains.

The Peñaflor site is located near Zaragoza on the left bank of the Gallego river, at an altitude of 300 mm above sea level. The landscape here is smooth and undulating and the Tertiary gypsiferous hills are covered by Plioquaternary glacial materials, composed of sands, clays and abundant calcareous stones. The soils at this site are deeper and are

classified as Calcisols. The site has a well developed forest of pine trees (*Pinus halepensis*). On the flatter areas, forest has been cleared for cultivation.

All the three study sites are almost level as they correspond to the tops of the plateau and hill landscapes of the central Ebro valley. The mean annual rainfall is 550 mm at Loma Negra, 450 mm at Valareña, and 300 mm at Peñaflor. The rainfall regime is similar in the three study sites. Rainfall is evenly distributed throughout the year around and is characterized by short and intense storm events.

Sample collection and analysis

At each of the three study sites, samples were collected from both uncultivated and cultivated land, i.e. two locations per site. The study sites were carefully selected to be reference sites. The uncultivated locations on communal rangeland were almost level and had undisturbed and stable soil covered by well preserved natural vegetation. The soil sampling at cultivated locations was established in nearby private farmlands that were also on flat terrain and had stable soils.

At each location, whole core samples were collected from a sampling grid comprising 9 points at Valareña and Peñaflor and 8 at Loma Negra due to the presence of rock outcrops. The sampling locations were of similar size with the sampling points spaced 1-2 m apart. Soil samples were collected using a 8.2 cm diameter corer. In order to examine the depth distribution of ^{137}Cs in the soil profile, the core collected from the centre of the grid network was sectioned into 5 cm depth increments. The depth of sampling was selected in order to include the entire ^{137}Cs depth profile and was 30 cm for the uncultivated land and 40 cm for the cultivated land.

A total of 52 samples were collected and these samples were air-dried and mill grinded to disaggregate the soil particles ensuring the break up of large soil aggregates. The individual samples were analysed to characterize their general properties, including: pH, organic matter (OM), electrical conductivity (EC), carbonate content and stoniness, i.e. the relative proportion of coarse fragments (> 2 mm) in the soil.

The pH (1:2.5 soil:water) and EC (1:5 soil:water) were measured using a pH-meter and a conductivity meter, respectively. Organic matter was determined by the Sanerlandt method¹³ using a titrimeter incorporating an ion selective electrode. Carbonates were measured using a pressure calcimeter¹⁴. The coarse fragments (> 2 mm) were separated from the fine fractions (< 2 mm) to what Cs-137 is strongly fixed. In order to separate the three grain-size fractions selected in this study: < 2 mm, 2-20 mm and > 20 mm, they were passed through 2 mm and 20 mm sieves.

A total of 264 samples, of which 46 were bulk samples and 42 sectioned samples, were split into 3 size fractions and prepared for Cs-137 analyses. Cs-137 was measured by gamma spectrometry at the Department of Geography, University of Exeter, UK, using hyperpure coaxial Ge detectors coupled to a multichannel analyser. The samples were loaded into Marinelli beakers for gamma assay at 662 keV and the counting times of 30000-60000 s provided an analytical precision of ca. ± 6 per cent at the 95% level of confidence. Cs-137 activity has been expressed per unit mass (Bq kg^{-1}) or per unit area as the inventory (Bq m^{-2}).

Results

Information on selected soil properties for the uncultivated and cultivated locations at the study sites are presented in Table 2. The soils have very low salinity values and they are alkaline with high carbonate percentages that increase from Loma Negra, through Valareña to the Peñaflor study site. At all the study sites, the soils have slightly higher organic matter contents at the uncultivated locations. As shown in Figure 2 stoniness is a main soil component at Peñaflor and Valareña and in relation with lithology the lower contents are in soils at Loma Negra. The mean value of stoniness is around 30% at the cultivated and uncultivated locations of Loma Negra, although the lowest and highest values are both found at the uncultivated location the mean stoniness percentage exceeds 40% at Valareña and Peñaflor and higher contents are found within the cultivated location at Peñaflor, whereas the contrary is observed at Valareña (Table 3). Summary statistics of the mass distribution in the different granulometrical fractions found at the uncultivated and cultivated sites of the study areas is presented in Table 4.

Previous research conducted in adjacent areas⁹ demonstrated that no ^{137}Cs fallout from the Chernobyl nuclear accident occurred at the study sites. The variability of the ^{137}Cs inventories at each study site has been analyzed. The particle size distribution and stoniness have been related to the distribution of ^{137}Cs inventories. As it can be seen in Figure 3, within the uncultivated location at the Valareña site, more than 94 % of the total ^{137}Cs inventory is contained in the < 2 mm fraction. Therefore, little ^{137}Cs is present in the coarser fractions and no ^{137}Cs was found in four of the nine samples.

At the Valareña cultivated location, the finer fraction accounts for 88% of the total ^{137}Cs inventory. The radioisotope was found in all samples of the 2-20 mm fraction, which

accounted for between 5 and 16 % of the total ^{137}Cs inventory. Low levels of ^{137}Cs were found in just one sample of the >20 mm fraction.

The ^{137}Cs inventories for the uncultivated locations show larger variations as indicated by the higher standard deviation. This conforms to previous reports of the higher variability found in rangelands¹⁵. In general, ^{137}Cs inventories are more homogeneous in cultivated areas, as indicated by the lower standard deviation (Table 5). This reflects the mixing effect of tillage that also has a strong effect on erosion in this area¹¹. Furthermore, cultivation causes the breaking down of the stones within the marls to smaller sizes. The importance of the coarser fractions, 2-20 and >20 mm, within the cultivated location is therefore smaller than for the uncultivated location. The largest difference is found in the 2-20 mm fraction that is less abundant at the cultivated location (see Table 4), and conversely the finer fraction is more abundant, thus increasing the mass of particles able to adsorb ^{137}Cs .

At Loma Negra, the proportion of the total ^{137}Cs inventory contained in the < 2 mm fractions is similar for both uncultivated and cultivated locations (around 95%). The average percentage of the total ^{137}Cs inventory in the 2-20 mm fractions at both the cultivated and uncultivated locations is around 4 %, which is lower than for the Valareña site. The content of ^{137}Cs of the >20 mm fractions at both the cultivated and uncultivated locations is below 0.5 %.

In Peñaflor almost the whole ^{137}Cs inventory is contained in the <2 mm fraction, and only three samples, two from the uncultivated location and one from the cultivated location, contain little ^{137}Cs in the coarse fractions.

The land use seems to affect most the ^{137}Cs reference inventories in soils on marls at Valareña and to a lower extent at Peñaflor, where higher values are found at the cultivated

locations. However, the effect of land use is almost negligible in soils on limestones at Loma Negra where ^{137}Cs inventories are almost equal at cultivated and uncultivated locations (Table 5). In most cases cultivated soils are not used as reference sites, however in mountain regions that are intensively cultivated it is difficult to find flat terrain with undisturbed and stable soil. Therefore, in spite of soil movement derived from tillage, flat cultivated soils developed on hard rocks such as limestones could be suitable sites to establish reference inventories.

As it can be seen from Figure 4, for the three study sites a direct and highly significant relationship exists between the ^{137}Cs inventories of the fine fraction and the total ^{137}Cs inventories ($r = 0.9$). However, the relationship was not significant when the ^{137}Cs inventories of the coarse fractions were related to total ^{137}Cs inventories, because most ^{137}Cs in the coarse fragments was found in soils on marls from Valareña.

Therefore, the type of lithology exerts an important control on the presence of ^{137}Cs in the coarse fractions. Thus, soft rocks, such as the marls and clays of Valareña are able to retain some ^{137}Cs in the larger particles. This could also be due to coating of larger particles with finer particles. Research on Australian soils has also found high percentages of ^{137}Cs in the gravel fraction¹². However, limestone which is the main component either as rock outcrop in Loma Negra, or as the sole stone component in Peñaflor does not contain ^{137}Cs .

For all the study sites, the lower values of standard deviation for the cultivated locations indicate that the ^{137}Cs inventories associated with the different grain size fractions are more homogenous as a result of the mixing effect of tillage. Furthermore, since in areas of high stoniness the content of the fine fractions that are able to retain the

radioisotope¹⁶ is lower, it would be preferable to sample larger areas, in order to obtain reliable ¹³⁷Cs inventories.

The different geographical location and altitude of the study areas affects the amount of total rainfall each site receives. There is a difference of around 100 mm per year between the three study sites. The highest annual rainfall is found at Loma Negra (500-550 mm), with lower values at Valareña (450 mm) and Peñaflor (300 mm). There is a clear relationship between the average ¹³⁷Cs inventories and the mean annual rainfall for the three study sites, as values increase from 1190, to 1500 and 1710 Bq m⁻² in Peñaflor, Valareña and Loma Negra, respectively (Figure 5). These differences therefore reflect the initial variation of ¹³⁷Cs fallout, which would increase in line with the increase in annual rainfall from Peñaflor to Loma Negra. The impact of the variation of annual rainfall on the ¹³⁷Cs inventories is more marked for uncultivated land than for cultivated land, since tillage homogenizes the radioisotope distribution within the soil.

Conclusions

To establish reliable reference inventories and to take account of the spatial variability of inventories found at reference sites in these semiarid environments, there is a need to include the 2-20 mm fraction in the ¹³⁷Cs analysis for soft rocks, such as marls, because the traditional procedure of sieving to < 2 mm could underestimate the inventory especially for cultivated soils. There appears to be no need to include the > 20 mm fraction for soils developed on hard rocks or containing limestone. Furthermore, because in semiarid mountain environments level and undisturbed sites are scarce the sampling of flat cultivated soils on hard rocks could be an alternative to establish suitable reference

inventories. These data may provide a basis for a better understanding of ^{137}Cs behaviour in these semiarid environments, with sparse vegetation cover and soils with a high stone content causing an increase in the variability of the initial spatial distribution of the radioisotope.

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References

- ¹R. J. LOUGHRAN, B. L. CAMPBELL, G.L. ELLIOT, Soil erosion in Agricultural Land, Ed. Wiley and Sons, Chichester, (1990) p. 119.
- ²J.C. RITCHIE, J. R. McHENRY, J. Environ. Qual., 9 (1990) 215.
- ³D. E. WALLING, T. A. QUINE, J. Soil Sci. 42 (1991) 147.
- ⁴J. C. RITCHIE, J. R. McHENRY, A. C. GILL, Ecology, 55 (1974) 887.
- ⁵D. E. WALLING, S. B. BRADLEY, C. J. WILKINSON, IAHS Publi. 159 (1986) 423.
- ⁶L. W. MARTZ, E. DE JONG, Catena, 14 (1987) 439.
- ⁷Y. N. BANIS, J. C. BATHURST, D. E. WALLING, Hydrol. Proc. 18 (2004) 795.
- ⁸C. PAPASTEFANO, J. Radioanal. Nucl. Chem. 267 (2006) 315.
- ⁹A. NAVAS, D. E. WALLING, IAHS Publ. 209 (1992) 129.
- ¹⁰A. NAVAS, J. MACHÍN, J. SOTO, Agr. Ecosyst.. Environ., 105 (2005) 493.

- ¹¹ T. QUINE, A. NAVAS, D. E. WALLING, J. MACHÍN, Land Degrad. Rehabil., 5 (1994) 41.
- ¹² D. J. MACFARLANE, R. J. LOUGHRAN, B. L.CAMPBELL, Aust. J. Soil Res. 30 (1992) 533.
- ¹³ F. GUITIAN, T. CARBALLAS, Técnicas de análisis de suelos, Ed. Pico Sacro, Santiago de Compostela, Spain, (1976) p. 288.
- ¹⁴ CSIC, Anal. Edafol. Agrobiol., 35 (1976) 813.
- ¹⁵ A. NAVAS, J. M. GARCIA-RUIZ, J. MACHÍN, T. LASANTA, D. E. WALLING, T. QUINE, B. VALERO, IAHS Publ., 245 (1997) 13.
- ¹⁶ J. SOTO, A. NAVAS, 2004. J. Arid Environ. 59 (2004) 719.

Figure captions

- 1.- The location of the study sites in the Ebro basin.
- 2.- The percentage of the total mass in the different grain size fractions at the uncultivated and cultivated locations at the study sites.
- 3.- The percentage of the total ^{137}Cs inventories accounted for by the different grain size fractions at the uncultivated and cultivated locations at Valareña and Loma Negra.
- 4.- Relationships between the total ^{137}Cs inventories and the ^{137}Cs inventories of the fine fraction and the coarse fractions for the study sites.
- 5.- Mean and standard deviations of ^{137}Cs inventories for the study sites.

Figure 1.

Figure 2

Figure 3.

Figure 4

Figure 5

Table 1. Main physiographic and land characteristics of the study sites.

Site	Climate	Landscape	Altitude (m)	Vegetation	Rainfall (mm)	Soil types	Parent material
Loma Negra	semiarid-temperate	plateaux	600	matorral, pines, cereals	500	Rendzic Leptosols	Limestones
Valareña	semiarid	plateaux	400	shrubs, cereals, fallow	450	Leptosols	marls, clays
Peñaflor	semiarid-arid	hills	300	pines, shrubs, cereals fallow	300	Calcisols	glacis: sands, limestones

Table 2. Soil properties at the study sites.

Studied area	Land use	pH	OM %	CO ₃ ⁼ %	Stoniness %	Electrical conductivity dSm ⁻¹
Loma Negra	cultivated	8.5	3.5	28.9	29.7	0.191
	uncultivated	8.5	4.1	23.9	28.3	0.186
Valareña	cultivated	8.3	4.8	35.1	38.0	0.306
	uncultivated	8.2	6.1	37.7	46.4	1.390
Peñaflor	cultivated	8.2	4.1	55.2	44.7	0.347
	uncultivated	8.5	4.9	42.5	39.6	0.193

Table 3. Basic statistics of the stoniness percentages for both the cultivated and uncultivated locations at the study sites a) and separately for the cultivated and uncultivated locations b).

a)

	<u>mean</u>	<u>sd.</u>	<u>min</u>	<u>max</u>
Peñaflor	42.1	4.6	30.4	49.6
Valareña	42.2	6.0	34.9	52.7
Loma Negra	29.0	9.2	9.4	46.5

b)

	Cultivated				Uncultivated			
	<u>mean</u>	<u>sd.</u>	<u>min</u>	<u>max</u>	<u>mean</u>	<u>sd.</u>	<u>min</u>	<u>max</u>
Peñaflor	44.7	3.5	40.4	49.6	39.6	4.3	30.4	43.9
Valareña	38.0	4.2	34.9	48.6	46.3	4.6	40.0	52.7
Loma Negra	29.7	3.4	24.6	33.4	28.3	13.0	9.4	46.5

Table 4. Basic statistics of the total mass distribution in the three grain size fractions at the studied sites.

<2 mm	Cultivated				Uncultivated			
	mean	sd.	min	max	mean	sd.	min	max
Peñaflor	55.3	3.5	50.4	59.6	60.5	4.3	56.1	69.6
Valareña	62.0	4.2	51.4	65.1	53.7	4.6	47.3	60.0
Loma Negra	70.3	3.4	66.6	75.4	71.7	13.0	53.5	90.6
2- 20 mm	mean	sd.	min	max	mean	sd.	min	max
	mean	sd.	min	max	mean	sd.	min	max
Peñaflor	31.1	6.4	24.8	44.6	26.3	4.7	15.8	30.1
Valareña	29.4	3.3	23.9	33.3	39.1	5.3	30.9	46.5
Loma Negra	18.2	3.2	13.2	22.4	14.4	7.1	6.7	25.4
>20 mm	mean	sd.	min	max	mean	sd.	min	max
	mean	sd.	min	max	mean	sd.	min	max
Peñaflor	13.6	7.5	3.3	24.8	13.2	5.3	6.7	21.6
Valareña	8.6	6.3	2.4	22.8	7.2	4.9	3.2	17.1
Loma Negra	11.5	3.1	6.9	17.9	13.9	6.9	2.7	21.1

Table 5.- Basic statistics of ^{137}Cs inventories (Bq m^{-2}) in the three grain size fractions at the cultivated and uncultivated locations at the study sites.

	Cultivated				Uncultivated			
	mean	sd.	min	max	mean	sd.	min	max
<2 mm								
Peñaflor	1194	188	939	1524	953	259	621	1437
Valareña	1574	266	1321	2203	1130	560	493	2421
Loma Negra	1604	271	1343	1991	1636	344	1115	2201
2- 20 mm								
Peñaflor	0	0	0	0	119	84	60	179
Valareña	200	73	88	289	62	87	0	213
Loma Negra	84	68	0	222	69	45	6	123
>20 mm								
Peñaflor	0	0	0	0	63	31	41	85
Valareña	8	25	0	74	24	35	0	79
Loma Negra	16	19	0	41	10	16	0	48
Total fractions								
Peñaflor	1194	188	939	1524	993	294	621	1437
Valareña	1783	274	1501	2469	1213	627	493	2712
Loma Negra	1698	305	1407	2213	1715	351	1130	2240