



^{137}Cs inventories along a climatic gradient in volcanic soils of Patagonia: Potential use for assessing medium term erosion processes



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ABSTRACT

Fallout radionuclides, such as Caesium-137, were proven to be a valuable means for studying medium-term (c.a. 50 years) soil erosion processes. In order to gain knowledge on the spatial distribution of ^{137}Cs in volcanic soils of Andean Patagonia, ^{137}Cs reference inventories were established along a precipitation gradient. At the subhumid sector, patterns of ^{137}Cs gain/loss associated with land use were also determined considering native forests under grazing, a degraded rangeland and a pine plantation.

Along the rising rainfall gradient (750–1400 mm), pedogenesis of volcanic ash soils, non-crystalline mineral formation, organic matter content and soil porosity varied, increasing in moister areas. Radionuclide inventories varied along the edaphoclimatic gradient, reaching mean values of 192, 267 and 576 Bq m⁻², at study areas with 750, 950 and 1400 mm of annual precipitation, respectively. The ^{137}Cs inventory followed an exponential relationship with precipitation, which could be related to the presence of allophane as the colloidal material in the soils from the rainiest area. The penetration depth reached by ^{137}Cs varied between 15 and 25 cm, according to rain amount and soil texture. Most of the ^{137}Cs fallout was retained in the uppermost 10 cm of the profiles and an exponential decline of ^{137}Cs with depth, highly related to organic matter contents, was found.

At the subhumid study area, both ^{137}Cs mass activity and inventory, significantly decreased under the different land uses, with respect to reference soils. Although the pattern of ^{137}Cs gain/loss varied according to topography, soil properties (organic matter and porosity) and vegetation cover, showing eroding and aggrading profiles, most samples had ^{137}Cs values lower than the reference value, suggesting loss of soil as a consequence of erosion processes. Furthermore, as much as 45%, 58% and 70% of sample points from native forests, plantation and rangeland, respectively, had ^{137}Cs values below the limit of detection. In the study transects, the loss of the upper 15 cm of soil in the subhumid sector during the last 50 years, highly exceeding tolerable erosion rates, highlights the urgent need for applying effective soil conservation measures.

Reference inventories, which vary according to the edaphoclimatic gradient, and the loss of the radionuclide in sites with anthropic intervention, show the potential for using ^{137}Cs measurements for assessing erosion processes in the Patagonian Andean Region.

1. Introduction

Soil erosion is a widespread problem throughout the world, frequently associated with land use changes, with negative impacts on sustainable production as well as on environmental conservation (e.g. Lal, 2017; Lizaga et al., 2018). Furthermore, land use changes have

strong impacts on variation of soil properties that occur in few decades (Navas et al., 2008; Lizaga et al., 2019). Patagonia is not outside of this global trend. The Patagonian Andean Region has suffered high anthropic pressure, associated with both the late nineteenth century period of white settlement and the earlier activities of the aboriginal population, who used fire for hunting guanacos (Veblen et al., 1999).

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Severe overgrazing by sheep and livestock during the early decades of twentieth century was the main factor in the shift in vegetation and soil degradation (Veblen and Lorenz, 1988). During the last century and at present, land use changes have triggered several soil erosion processes, being the region subjected to deforestation, grazing, fires, agriculture, exotic plantations, and urbanization (Carabelli and Scoz, 2016; Colazo et al., 2019). Recent studies showed that the replacement of native forest modified physical, chemical and magnetic soil properties, resulting from soil desiccation and led to selective removal of fine particles by erosion (La Manna et al., 2018a). Volcanic soils, dominant in this region, were highly erodible where the mineral soil remained exposed, without litter or vegetation cover (La Manna et al., 2016).

In order to understand the influence of land use on soil degradation and to select effective soil conservation measures, reliable quantification of soil erosion rates is a requirement (Navas et al., 2014). Isotopic techniques based on the use of fallout radionuclides such as Caesium-137 (^{137}Cs), affords a valuable means for studying erosion and deposition processes within the landscape and have been increasingly applied for quantifying medium-term (c.a. 50 years) soil erosion rates, mainly in the northern hemisphere. ^{137}Cs was introduced into the atmosphere by the detonation of fission nuclear weapons. Its use for erosion studies is based on the strong fixation of the radionuclide by the fine components of the soil (Tamura, 1964) and its subsequent redistribution in the landscape by physical processes. Although the global pattern of ^{137}Cs inventories shows a latitudinal variation, with much higher values in the northern hemisphere, the ^{137}Cs input showed that South America would be a region appropriate for soil erosion studies using the radiometric technique (García Agudo, 1998), though it is required a ^{137}Cs deposition in the soil great enough to allow its precise quantification. About 25% of the total global fallout has been deposited in the southern hemisphere (UNSCEAR, 1982; UNEP, 1984). ^{137}Cs deposition not only depends on the latitude, but also on the amount of rainfall, since the radionuclide is removed from the atmosphere by rain, snow or dry precipitation (García Agudo, 1998). Several studies have shown that ^{137}Cs deposition is positively correlated with precipitation (Pálsson et al., 2006; Porto et al., 2011). This has been shown along a precipitation-altitudinal gradient from semiarid to alpine environments in N Spain (Navas et al., 2007). Furthermore, physiographic conditions with effect on local rainfall patterns also have an effect on the initial spatial distribution of the ^{137}Cs fallout (Porto et al., 2001).

The potential of ^{137}Cs as tracer of soil erosion has been successfully demonstrated in a wide range of environments (Ritchie et al., 1970; Walling et al., 1986; Pennock and de Jong, 1987; Zhang et al., 1990; Navas and Walling, 1992; Owens et al., 1996; Sadiki et al., 2007; Gaspar et al., 2013; Quijano et al., 2016a). The results of these studies show that the spatial distribution of ^{137}Cs reflects soil redistribution processes and sediment transport, which are controlled by topographic and land use conditions. However, this technique rarely has been used in South America. In Argentina, Buján et al. (2000) and Juri Ayub et al. (2008, 2010) applied fallout ^{137}Cs in the relatively flat central areas of Cuyo and Pampean regions, but no published information is available from the Patagonian Andean Region. The nearest studies were carried out in Chile, at the east from Andes mountain chain, with different edaphoclimatic conditions (Schuller et al., 1997, 2002).

Soils of the Patagonian Andean Region are developed mainly from volcanic ashes and, even worldwide, studies about ^{137}Cs in volcanic soils are rare (Cox and Fankhauser, 1984; Schuller et al., 2002; Sigurgeirsson et al., 2005; Muñoz-Salinas and Castillo, 2018). To fill this gap studies aimed at improving the knowledge on the ^{137}Cs behaviour in volcanic soils are needed, mainly studies in relation to erosion processes, since volcanic soils are highly erodible where the soil remains bare (La Manna et al., 2016). Studies about ^{137}Cs in volcanic soils showed that common soil conservation practice, such as ditches, trenches and afforestation, could result non effective in these soils (Muñoz-Salinas and Castillo, 2018).

Soils formed over volcanic ashes have, as dominant clays, non-

crystalline minerals (e.g., allophane, imogolite) which give distinctive properties to soil (Dahlgren et al., 2004; McDaniel et al., 2012). Caesium-137 has a very short radius of hydration, and it is strongly adsorbed to cation exchange sites of colloids (Tamura, 1964). While clays are the most important binding agent of ^{137}Cs in mineral soils (Ritchie et al., 1970), the accumulation of organic matter, typical of volcanic soils, may improve ^{137}Cs retention (Sigurgeirsson et al., 2005) as it has been shown the key role of organic matter in fixing the radionuclide (Gaspar et al., 2013). Several physical and chemical soil properties control ^{137}Cs behaviour, such as type of clay minerals, pH, cation exchange capacity (CEC), organic carbon content and texture (Gaspar et al., 2013; Quijano et al., 2016b). Studies on volcanic soils showed that ^{137}Cs is strongly retained in the uppermost few centimetres of soil, by non-crystalline minerals, and that the vertical migration of the radionuclide is controlled by clay content and porosity (Cox and Fankhauser, 1984; Schuller et al., 2002; Sigurgeirsson et al., 2005).

The landscape and soils in the Patagonian Andean region are characterized by a great edaphoclimatic gradient in a short distance (Colmet Daage et al., 1988). The abrupt east-west pluviometric gradient, evidenced by vegetation which changes from forests at the west to steppe towards the east, also controls the pedological processes suffered by volcanic ash (Parfitt et al., 1984). Mineralogical studies support that in the west, allophane is formed, whereas to the east—subhumid sector—soils are enriched in imogolite (non-crystalline) or/and halloysite (1:1 layer silicate) (Besoin, 1985; Colmet Daage et al., 1988). Thus, physical and chemical properties greatly varied in the east-west gradient (Gaitán and López, 2007), which could affect ^{137}Cs behaviour (Tamura, 1964; Gaspar and Navas, 2013). Research done in volcanic soils of Iceland showed that ^{137}Cs was strongly retained by allophane (Sigurgeirsson et al., 2005).

Our study aims to gain knowledge on the spatial distribution of ^{137}Cs in volcanic soils of Andean Patagonia, by measuring reference inventories along an edaphoclimatic gradient and to analyse the factors controlling the variation of ^{137}Cs mass activities and inventories. We pursue to determine the vertical distribution of the radionuclide under representative land covers and land uses in the region and compare ^{137}Cs patterns of gain and loss associated with different land uses in the subhumid sector, considering the three most representative ones in the region: native forests under grazing, degraded rangeland and pine plantation.

2. Materials and methods

The study was carried out along a longitudinal gradient, at ca. 43° south latitude, from the forest-steppe ecotone in the subhumid sector, up to the Patagonian Andean forests towards the west in Argentina, comprising a mean annual precipitation gradient from 750 mm to 1400 mm, and an edaphic gradient from volcanic soils without non-crystalline minerals in the subhumid sector to allophanized soils in the rainiest area.

Along this gradient, three study areas were selected: Esquel (71°23' WL; 42°56' SL; 750 mm mean annual precipitation), Trevelin (71°28' WL; 43°2' SL; 950 mm) and Los Alerces National Park (71°45' WL; 43°10' SL; 1400 mm) (Fig. 1). A pristine, flat and stable site, which has suffered neither erosion nor soil accumulation, was selected for establishing the ^{137}Cs reference inventory in each of the three study areas. In each reference site, sampling points, separated ca. 20–50 m, were selected. We collected, by using a 5.9 diameter cylinder, depth interval soil profiles, where samples were sectioned at 5 cm depth increments, and soil bulk samples. In Esquel, seven points were sampled; of these, six were sectioned at 5 cm depth increments. In Trevelin, four points were sampled; two of which were sectioned. In Los Alerces National Park, five points were sampled; three of which were sectioned. To retain the entire ^{137}Cs profile and taking into account the probable penetration depth of the radionuclide in the region, samples were collected until 25 cm depth in Esquel and Trevelin and until 30 cm depth



Fig. 1. Location of the study area.

in Los Alerces National Park. A pit was dug in each study area, and soil morphological characteristics were recorded according to USDA (Schoeneberger et al., 1998).

In Esquel, the eastern part of the study area, different land uses were also evaluated. Soil samples were taken from two native forest patches, as well as from transects along a pine plantation and a rangeland. Native forests, dominated by *Maytenus boaria* Mol with a shrub stratum, are preserved as isolated patches in an anthropized area, with different degradation conditions. Forest A, currently is a forest patch with close crowns, but in aerial photographs from 1970 it appears as a bushy patch, with only a few high crowns (Fig. 2a). Forest B, both at 1970 as at present, corresponds to a small patch of trees in a degraded and eroded area (Fig. 2b). Both studied forests are grazed and used as shelter by the cattle. Sampling was carried out in microsites where, currently, the soil is completely covered by litter. Eleven sampling points were considered, including five sectioned at 5 cm depth increments and six bulk samples.

The afforestation (pine plantation) and the rangeland transects were located along two adjacent slopes. The rangeland, used for grazing for the last 100 years, presented grass shrub vegetation (80% covered),

dominated by *Rumex acetosella* L., a perennial and invasive exotic herbaceous plant, accompanied by species indicative of degradation and overgrazing such as *Acaena splendens* Hook. & Arn and *Mulinum spinosum* (Cav.) Pers. and grasses of low palatability such as *Pappostipa speciosa* Trin. et Rupr. At the upper part of the slope transect, the rangeland presents native trees of *Maytenus boaria*.

The afforestation corresponded to a 32 years *Pinus radiata* D. Don plantation from 1985, and a *Pinus ponderosa* plantation from 1996 at the upper part of the slope. At the uppermost slope, patches of native vegetation, mainly shrubs, are intermixed with the pines (Fig. 2c).

Since the rangeland and the afforestation are located on hill slopes, the sampling was conducted along a transect parallel to the main slope direction. Ten and twelve sampling points placed ca. 25 m apart from each other, were sampled along the transects of the rangeland and plantation (Fig. 2c). At each sampling point, a bulk sample was collected at 0–25 cm depth, and in three points (the upper, middle and lower slope) soil samples were sectioned at 5 cm depth increments. Where deposition was supposed to happen, sampling depth was extended until 40 cm depth to ensure that the entire ¹³⁷Cs profile was retained.

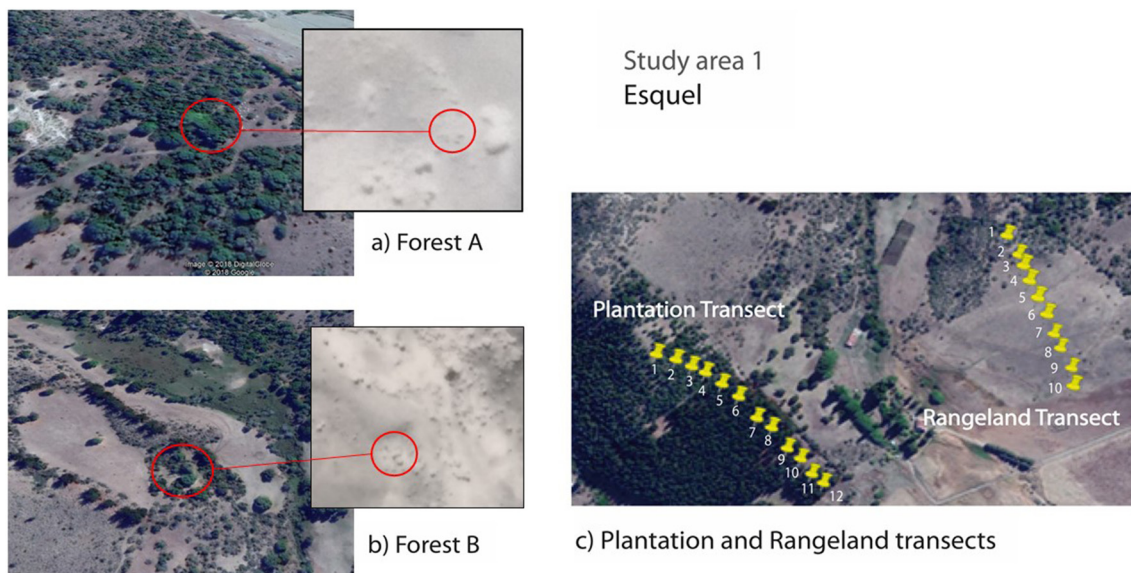


Fig. 2. Location of the soil profiles sampled at the study site 1 - Esquel: a) Forest A, b) Forest B and c) along two transects.

The microtopography of the slopes was evaluated by geometric levelling, using a TOPCON AT-G6 optical level and a 1 cm-graduated telescopic sight.

All the soil samples were air-dried, homogenised and passed through a 2 mm for radionuclide gamma assays and physical and chemical properties analyses. The mass activity of ^{137}Cs was measured using a Canberra high-resolution, low-background, hyperpure germanium coaxial gamma detector coupled to an amplifier and multichannel analyser. The detector had a relative efficiency of 50% and a resolution of 1.9 keV (shielded to reduce background), and was calibrated using standard certified soil samples that had the same geometry as the measured samples. Subsamples of 50 g were loaded into plastic containers. Count times over 24 h provided an analytical precision of about $\pm 2\text{--}3\%$ at the 95% confidence level. Mass activities were expressed as Bq kg^{-1} dry soil. Inventories were expressed as activity per unit area (Bq m^{-2}) which was calculated using the weight of the < 2 mm fraction as the radionuclide is only fixed in the fine fraction of the soil (Soto and Navas, 2004) and the cross section of the sample (Gaspar and Navas, 2013). The gain/loss soil under different land uses was calculated in each sample point from the difference between ^{137}Cs remaining in the profile and the ^{137}Cs reference level (Navas et al., 2013).

The soil texture was analyzed by Coulter laser granulometer after destruction of organic matter with 10% H_2O_2 at 80 °C. Organic matter (SOM) was analyzed by loss on ignition (IRAM-SAGPyA, 2008). Bulk density was determined in undisturbed samples, and total porosity was calculated (Taboada and Alvarez, 2008). The pH in NaF 1N (1:50) was measured using pH-meter. pHNaF measured at 2 and 60 min is considered an indicator of non-crystalline aluminosilicates in soils (Fiedes and Perrot, 1966).

Differences in soil properties between the study sites along the longitudinal gradient were analyzed by two-way analysis of variance (ANOVA), considering the study site (3 levels) as factor. Tukey's HSD (Honestly Significant Difference) was applied as post hoc analysis when ANOVA test showed a p -value < 0.05 . The relationship between the ^{137}Cs inventory and precipitation rate was analyzed by linear and non-linear regression models, selecting the model with the best fit. Correlation between ^{137}Cs mass activity and soil properties was evaluated by Pearson test.

Vertical distribution of ^{137}Cs was evaluated and the correlation between ^{137}Cs activity and organic matter content along the soil profiles was evaluated, separately, for reference sites and for the different land uses in Esquel, considering the sectioned samples.

Differences in soil properties between the different land uses at Esquel were analyzed by two-way analysis of variance (ANOVA) and Tukey's HSD. The correlation between ^{137}Cs activity and soil properties was also evaluated by Pearson test, considering the bulk samples (0–25 cm) along the slope.

The statistical analyses were carried out with the Infostat software (Di Rienzo et al., 2013).

3. Results

3.1. Reference ^{137}Cs inventory

Table 1 shows morphological features of representative soils in the study area, where Holocene volcanic ash was the main soil parent material. Soils resulted light loose and easily excavated and allowed roots to penetrate to great depths. Along the precipitation gradient, soils were well drained, with medium and coarse textures. In Esquel and Los Alerces National Park, soils showed scarce horizons development, whereas in Trevelin, with finer textured soils, a Bw horizon was recorded.

Along the precipitation gradient, physical and chemical soil properties greatly varied, mainly by the presence of non-crystalline aluminosilicates, which was also related to organic matter content and bulk density (Table 2). The Los Alerces National Park was the only site where

soils were rich in allophane. All the soil samples from Los Alerces showed high pH NaF values (i.e., pH NaF 2' > 9.2), suggesting the presence of allophane (Irisarri, 2000). At Trevelin, soils were rich in imogolite (i.e., pH NaF 60' > 9.2) and at Esquel most samples showed absence of non-crystalline minerals. The organic matter contents increased along the edaphoclimatic gradient. The stable aggregation of volcanic soils resulted in low density and high porosity, the last one increasing in allophanized soils (Table 2).

Textural classes varied from loam to sand, being representatives of dominant soil textures from the study area (La Manna et al., 2018b). In Esquel, soil samples were classified as sandy loam, loamy sand and sand; in Trevelin textures were loam and silt loam; while in Los Alerces National Park textures varied from silt loam to sandy loam.

The reference ^{137}Cs activity and ^{137}Cs inventory both increased progressively in relation to the rising rainfall gradient, reaching in Los Alerces National Park, values that almost were three times higher than at Esquel (Table 3). The ^{137}Cs inventory was positively correlated with the mean annual precipitation rate, following an exponential relationship, as shown in Fig. 3. The allophanic soils of Los Alerces National Park, even having low clay contents (Table 2), showed ^{137}Cs values even higher than the expected by a lineal regression model between ^{137}Cs inventories and annual precipitation (Fig. 3).

In the study area, ^{137}Cs was positively associated not only with precipitation but also with pH NaF ($r = 0.53$; $p = 0.03$), organic matter ($r = 0.45$; $p = 0.08$) and soil porosity ($r = 0.51$; $p = 0.04$), soil variables increasing in relation to precipitation rates (Table 2).

3.2. Vertical distribution of ^{137}Cs

In all the study areas, most of ^{137}Cs fallout was retained in the uppermost 10 cm of the soil. As much as 95, 90 and 87% of ^{137}Cs was found in the 0–10 cm depth interval in Esquel, Trevelin and Los Alerces National Park, respectively. An exponential decline of ^{137}Cs with depth was found (Fig. 4). However, in Esquel, some profiles showed the highest ^{137}Cs values at 5–10 cm depth, instead of at 0–5 cm.

The ^{137}Cs activity along the soil profiles varied according to organic matter content. Organic matter along the profiles followed the same pattern as ^{137}Cs for the three study areas, showing significant correlation between ^{137}Cs activity and organic matter: $r = 0.81$ ($p < 0.001$); $r = 0.86$ ($p = 0.01$); $r = 0.93$ ($p < 0.001$) for Esquel, Trevelin and Los Alerces National Park, respectively.

The penetration depth of ^{137}Cs varied between the study sites. The greatest penetration was 25 cm in Los Alerces National Park, whereas in Esquel and Trevelin the radionuclide was detected just up to 15 cm depth, with the exception of one site from Esquel, which showed a penetration of 20 cm depth.

3.3. ^{137}Cs in Esquel: relationship with land use

Soil properties varied according to the land use. Native forest showed the greatest values of pH NaF, organic matter, porosity and silt granulometric fraction and the lower sand proportion (Table 4). Unlike the other study sites in Esquel, some soils under native forest presented non-crystalline minerals (imogolite), which even differed from reference soils (see Table 2).

Despite these differences in physical and chemical properties, the values of ^{137}Cs , both mass activity and inventory, did not differ between the different land uses (Table 5). However, ^{137}Cs was significantly lower ($p < 0.05$) in soils from native forest under grazing, pine plantation and rangeland, than in reference sites (see Table 2).

3.3.1. Native forests

The ^{137}Cs was totally lost in some samples by erosion, whereas in other points, the ^{137}Cs inventories increased reaching values higher than those found in references, indicating soil accumulation from elsewhere (Fig. 5). The loss of ^{137}Cs , compared to reference inventory,

Table 1
Morphological soil properties of the reference sites along a precipitation gradient in the Patagonian Andean region.

Horizon	Depth (cm)	Color ^a	Texture ^b	Structure ^c	Roots ^d	Rock fragments (%)	NaF reaction	Boundary ^e
Sampling site 1 - Esquel - precipitation: 750 mm - Mollisol								
A	0–25	10YR 3/3	Ls	1 SBK F	2 F	15	–	C W
AC	25–70	10YR 3/4	Ls	1 SBK M	2 F	15	–	C W
C	70–90+	10YR 3/4	S	1 SBK CO	2 M	20	–	
Sampling site 2 - Trevelin - precipitation: 950 mm - Andisol								
A	0–23	10YR 3/2	Ls	2 GR F	2 F	0	+	C S
ABw	23–48	10YR 4/3	Ls	1 SBK M	3 M	0	+	G S
Bw	48–75	10YR 4/4	L	1 GR M	2 CO	10	+	C W
C	75–110+	10YR 5/4	scl GX	0 MA	1 CO	70	+	
Sampling site 3 - Los Alerces National Park - precipitation: 1400 mm - Andisol								
A	0–16	10YR 3/2	Sl	0 SGR	3 F	0	+	C W
AC	16–45	10YR 3/4	Ls	0 SGR	3 CO	0	+	C S
C1	45–75	10YR 4/4	S	0 MA	1 M	0	+	G S
C2	75–125+	10YR 5/4	S	0 MA	1 M	0	+	

^a Color: Colors are under moist condition according to Munsell soil color charts (1990).

^b Texture: L = loam, S = sand, Ls = loamy sand, Sl = sandy loam, scl = sandy clay loam. GX = extremely gravelly.

^c Structure: 0 = structureless, 1 = weak, 2 = moderate; MA = massive, GR = granular, SGR = single grain, SBK = subangular blocky; F = fine, M = medium, CO: coarse.

^d Roots: 1 = few, 2 = common, 3 = many; F = fine, M = medium, CO = coarse.

^e Horizon boundary: A = abrupt, C = clear, G = gradual, S = smooth, W = wavy.

Table 2
Physical and chemical soil properties of the reference sites along a precipitation gradient in the Patagonian Andean region. Mean values and standard errors for 0–25 cm depth are shown. Different letters indicate significant differences between the study areas.

	Esquel	Trevelin	Los Alerces National Park
Precipitation (mm)	750	950	1400
pH NaF 2'	8.0 ± 0.1 c	8.6 ± 0.3 b	10.3 ± 0.03 a
pH NaF 60'	8.8 ± 0.2 c	9.5 ± 0.3 b	11.2 ± 0.04 a
Non crystalline minerals	–	Imogolite	Allophane
SOM (%)	8.8 ± 1.1 b	13.4 ± 1.5 a	14.0 ± 0.2 a
Bulk density (gcm ⁻³)	0.82 ± 0.11 a	0.72 ± 0.07 ab	0.63 ± 0.03 b
Total porosity (%)	69.1 ± 4.0 a	72.8 ± 2.5 ab	76.2 ± 1.0 b
Clay (%)	4.7 ± 0.8 b	8.6 ± 0.9 a	4.7 ± 0.3 b
Silt (%)	29.2 ± 4.0 b	52.2 ± 1.8 a	46.9 ± 1.3 a
Sand (%)	66.1 ± 4.6 a	39.2 ± 1.9 b	48.4 ± 1.5 b

varied between 27 and 100% and 89 and 100% for Forest A and B, respectively. While the gain of ¹³⁷Cs showed a maximum value of 19% in Forest A and 9% in Forest B. These results suggest a net loss of soil in these patches of native forests. Erosion process seems to be more severe in Forest B, which appears as a small patch of trees in a degraded and eroded area (Fig. 2b).

The ¹³⁷Cs mass activity was significantly correlated with porosity ($r = 0.69$; $p = 0.03$) and tended to be related with organic matter content ($r = 0.45$; $p = 0.10$).

The vertical distribution of ¹³⁷Cs was similar to the found in reference sites, showing an exponential decline with depth (Fig. 6). The penetration depth of ¹³⁷Cs was 20 cm and 10 cm for Forest A and B, respectively. A significant correlation was found between ¹³⁷Cs activity and organic matter content (Person's $r = 0.67$; $p < 0.01$).

3.3.2. Plantation and rangeland

In the highest part of the pine plantation hillside, the ¹³⁷Cs inventory was similar to those found at the Esquel reference sites (P1 in Fig. 7). However, as seen in Fig. 7, there were marked differences along the plantation transect, and there was erosion in most sample points. The radionuclide was not detected in many samples. On the other hand, at the bottom slope and in sectors along the hill where the slope degree decreases forming micro depressions, the percentage of ¹³⁷Cs loss

Table 3
¹³⁷Cs inventories and mass activities in soils from the Patagonia Andean region. Values for 0–25 depth are shown. Different letters indicate significant differences between the study areas.

	¹³⁷ Cs Inventory (Bq m ⁻²)		
	Esquel	Trevelin	Los Alerces National Park
n	7	4	5
Mean	192.25 a	266.71 b	576.08 c
Median	189.23	268.99	579.26
Standard deviation	18.16	22.06	38.28
Standard error	6.86	11.03	17.12
Value range	174.62–219.39	240.89–287.99	523.92–620.44
	¹³⁷ Cs Activity (Bq Kg ⁻¹)		
	Esquel	Trevelin	Los Alerces National Park
n	7	4	5
Mean	6.22 a	9.17 b	19.72 c
Median	5.45	9.17	19.72
Standard deviation	2.17	1.13	1.19
Standard error	0.89	0.80	0.84
Value range	3.40–8.84	8.37–9.97	18.88–20.56

varied between 0.2 and 54%.

The ¹³⁷Cs mass activity for the bulk samples (0–25 cm depth) was significantly correlated with organic matter content ($r = 0.61$; $p = 0.03$) and porosity ($r = 0.64$; $p = 0.02$).

Along the rangeland transect, most samples (7 out of 10) showed negligible values of the radionuclide, that was below the limit of detection (Fig. 8). However at the bottom slope (P10 in Fig. 8) and at the uppermost slope (P3), where soil is partially protected by shrubs and native trees (Fig. 2c), soil loss was lower, ca. a loss of 35–41% of the ¹³⁷Cs reference inventory. In the highest part of the hillside (P1), radionuclide gain was recorded, with ¹³⁷Cs inventory around 13% higher than the reference value (Fig. 8).

The ¹³⁷Cs activity for the bulk samples (0–25 cm depth) was significantly correlated with porosity ($r = 0.84$; $p = 0.02$) and tended to be positively related to clay content ($r = 0.53$; $p = 0.10$).

Along both plantation and rangeland transects, an exponential

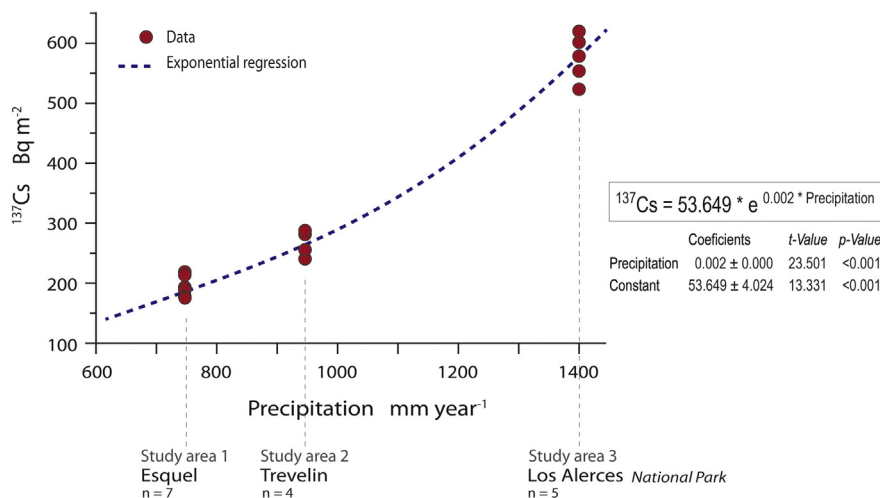


Fig. 3. Relationship between ¹³⁷Cs inventory (Bq m⁻²) and precipitation for the study site 1 - Esquel, study site 2 - Trevelin and study site 3 - Los Alerces National Park.

decline of ¹³⁷Cs with depth was found in the profiles. The maximum depth reached by ¹³⁷Cs in the plantation was 10 cm in the profile from the highest part of the hillside (P1), whereas in P3 and P12, ¹³⁷Cs was found only in 0–5 cm depth samples (Fig. 7). On the other hand, the depth reached by ¹³⁷Cs in the rangeland was 15 cm in the profile from the highest part of the hillside (P1) and 10 cm in P3 and P10 (Fig. 8).

The radionuclide pattern in the soil profiles was significantly related to organic matter content: Pearson's $r = 0.85$ ($p < 0.001$); $r = 0.63$ ($p = 0.04$), for plantation and rangeland, respectively.

4. Discussion

4.1. Reference ¹³⁷Cs inventory

The edaphoclimatic gradient, characteristic of the Patagonian Andean Region (Colmet Daage et al., 1988), was covered in this study, from soils without non-crystalline aluminosilicates in Esquel, soils with imogolite in Trevelin, to allophanized soils in Los Alerces National Park (Table 2). In agreement with increasing in pH NaF (indicator of non-crystalline aluminosilicates) according to precipitation rates, organic matter and soil porosity also increased towards the west.

Allophane and imogolite, under an electron microscope, appear as nanoparticles of hollow spheres and hollow threads or tubules,

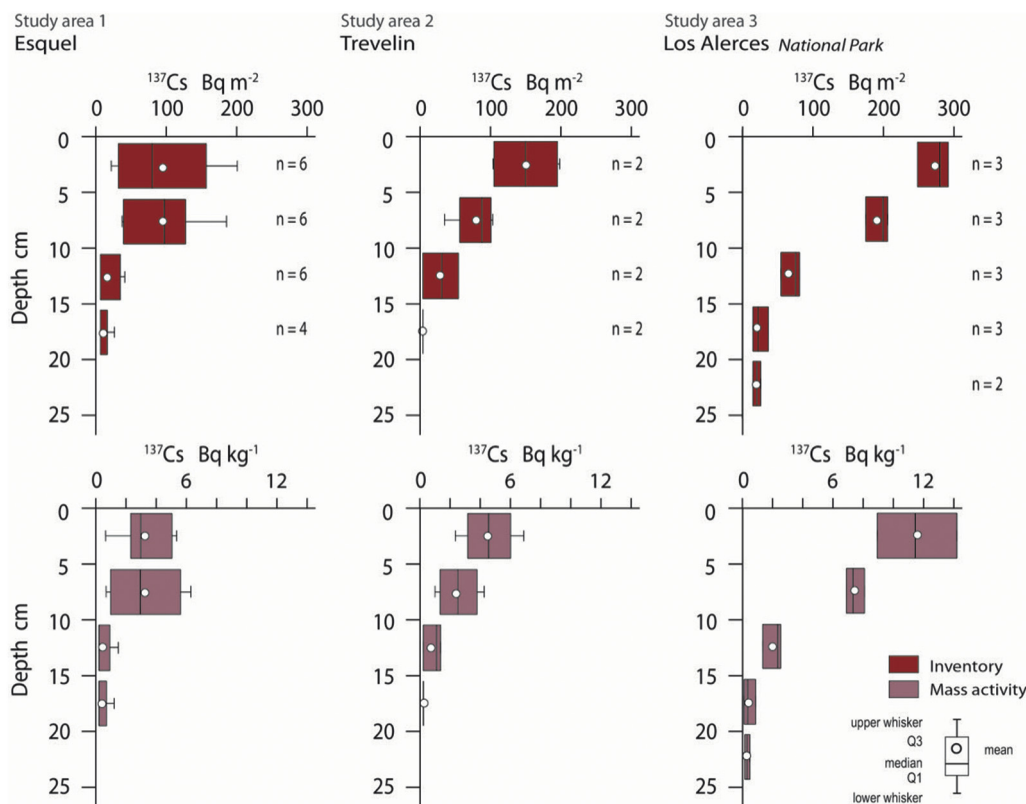


Fig. 4. Mean values of ¹³⁷Cs mass activity (Bq kg⁻¹) and Inventory (Bq m⁻²) for sectioned soil profiles at the study site 1 - Esquel, study site 2 - Trevelin and study site 3 - Los Alerces National Park.

Table 4

Physical and chemical soil properties under different land uses at the study site 1 - Esquel. The properties are summarised for 0-25 cm depth. Mean values and standard errors are shown. Different letters indicate significant differences between land uses.

	Native forest	Plantation	Rangeland
pH NaF 2'	8.5 ± 0.1 b	8.0 ± 0.1 a	7.9 ± 0.1 a
pH NaF 60'	9.0 ± 0.1 b	8.6 ± 0.1 a	8.5 ± 0.1 a
Non crystalline minerals	None - Imogolite	-	-
SOM (%)	13.5 ± 1.2 b	4.2 ± 0.3 a	4.8 ± 0.2 a
Bulk density (g cm ⁻³)	0.74 ± 0.11 a	1.10 ± 0.09 b	1.09 ± 0.05 b
Total porosity (%)	72.0 ± 4.0 b	58.4 ± 3.5 a	58.9 ± 1.8 a
Clay (%)	4.4 ± 0.5 ab	3.2 ± 0.6 a	5.9 ± 0.6 b
Silt (%)	44.9 ± 4.5 b	16.8 ± 2.8 a	27.3 ± 2.2 a
Sand (%)	50.7 ± 4.6 a	80.0 ± 3.3 c	66.8 ± 2.7 b

Table 5

¹³⁷Cs inventories and mass activities in soils under different land uses at the study site 1 - Esquel. Values are summarised for 0-25 cm depth. Mean values and standard errors are shown. Same letters indicate non-significant differences between land uses.

¹³⁷ Cs Inventory (Bq m ⁻²)			
	Native forest	Plantation	Rangeland
n	11	12	10
Mean	75.95 ± 29.18 a	54.14 ± 23.22a	46.00 ± 24.89a
Median	21.15	0	0
Value range	0-229.03	0-193.61	0-217.98
¹³⁷ Cs activity (Bq Kg ⁻¹)			
	Native forest	Plantation	Rangeland
n	11	12	10
Mean	2.21 ± 0.96a	0.86 ± 0.37a	1.06 ± 0.57 a
Median	0.41	0.19	0
Value range	0-8.36	0-3.40	0-4.83

respectively (Theng and Yuan, 2008). Unique physical attributes of volcanic soils are related to structural assemblages of these hollow spheres and tubules as mineral entities into resilient, progressively larger aggregated domains (McDaniel et al., 2012). This stable aggregation results in low density, high porosity and high surface area, as it was found in the study area (Table 2). According to these properties, cation exchange capacity of volcanic soils is relatively high, often higher than 50 cmolc kg⁻¹ (McDaniel et al., 2012). Soil surveys developed in the study area found CEC higher than 100 cmolc kg⁻¹ in allophanized soils, even in coarse textured soils (Morales and La Manna,

2011).

The ¹³⁷Cs reference inventory varied from 192 to 576 Bq m⁻² (Table 3), and was positively related to the mean annual precipitation rate, in accordance with several studies both in volcanic (Cox and Fankhauser, 1984; Schuller et al., 2002; Sigurgeirsson et al., 2005) and non-volcanic soils (e.g. Blagoeva and Zikovskiy, 1995; Pálsson et al., 2006; Navas et al., 2007; Porto et al., 2011). It is known that atmospheric washout, and therefore precipitation, is one of the dominant factors controlling the deposit of radionuclides. The highest values of ¹³⁷Cs inventory (576 Bq m⁻²) were found in Andisols from Los Alerces National Park, with 1400 mm of annual precipitation rate, which resulted almost three times higher than those found in Esquel. Studies on volcanic soils from Chile, located at northern latitudes than our study area (38°-41° South Latitude), recorded ¹³⁷Cs inventories from 450 to 5410 Bq m⁻², highly correlated with annual precipitation rates, which ranged from 750 mm to 4000 mm (Schuller et al., 2002).

The relationship between ¹³⁷Cs inventory and precipitation followed an exponential model (Fig. 3); showing the allophanic soils from the rainiest sector, ¹³⁷Cs values higher than the expected by a lineal regression model. This result could be related to the presence of allophane as colloidal material in Los Alerces National Park. Studies on volcanic soils of Iceland showed that ¹³⁷Cs fallout is strongly retained by allophane (Sigurgeirsson et al., 2005). Thus, in the studied rainfall gradient, ¹³⁷Cs in soils was not only related with annual precipitation, but also with key soil properties such as pH NaF, organic matter and porosity, which are soil properties that vary according to pedological processes suffered by ash in relation to the amount of precipitation (Parfitt et al., 1984).

4.2. Vertical distribution of ¹³⁷Cs

The penetration depth reached by ¹³⁷Cs varied in the study area, according to rain amount and soil characteristics, reaching maximum values of 20, 15 and 25 cm for Esquel, Trevelin and Los Alerces National Park, respectively. The maximum depth reached by ¹³⁷Cs along the edaphoclimatic transect could be explained by mean annual precipitation and by soil textures, since high rainfall rates (as in Los Alerces National Park) and coarse textures (as those found in Esquel) favour downward migration of the radionuclide (Schuller et al., 1997). In Trevelin, where soils showed the finest textures with clay contents significantly higher than the others study sites (Table 2), penetration depth was similar or even lower than in Esquel, in spite of increasing precipitation. Several studies show that precipitation, together with texture and coarse-pore volume control the vertical migration of ¹³⁷Cs (He and Walling, 1996). The penetration depth of ¹³⁷Cs found in Los Alerces National Park (i.e., 25 cm) agrees with results found by Schuller et al. (2002) in volcanic soils from Chile with similar precipitation, but

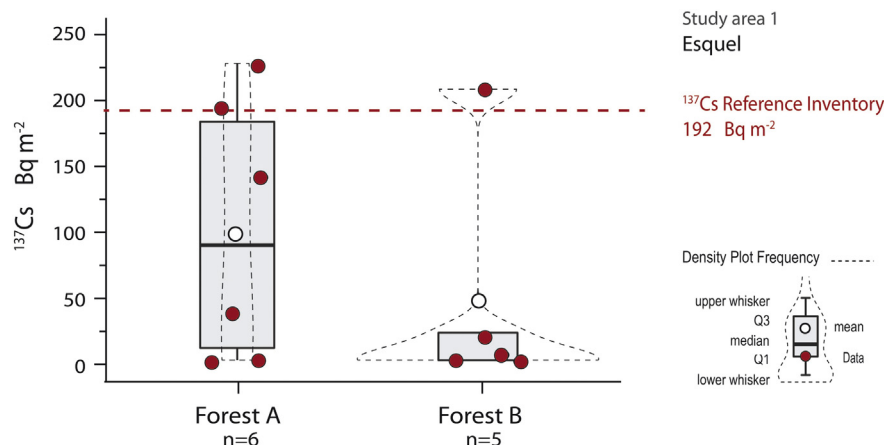


Fig. 5. Inventories of ¹³⁷Cs (Bq m⁻²) for soil profiles in forest A and forest B at the study site 1 - Esquel.

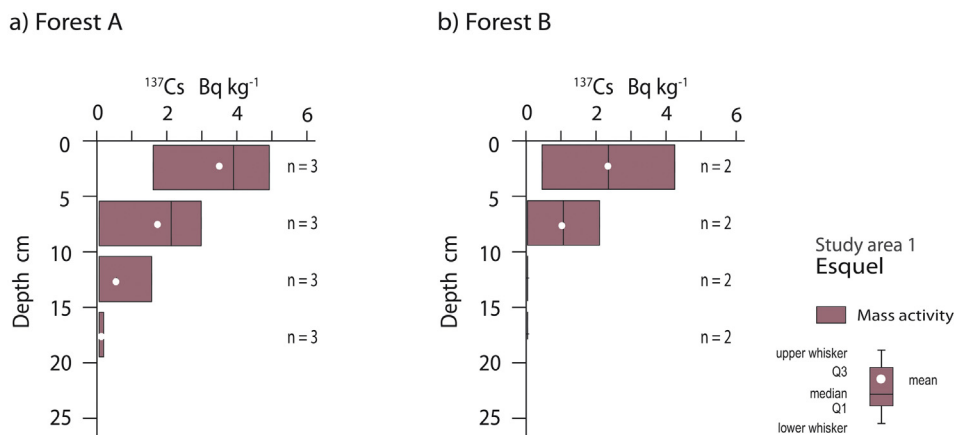


Fig. 6. Mean values of ¹³⁷Cs mass activity (Bq kg⁻¹) for sectioned soil profiles at the study site 1 - Esquel in forest A and forest B.

reaching much higher values, up to 70 cm, where mean precipitation increases up to 4000 mm.

Most of the ¹³⁷Cs fallout was retained in the uppermost 10 cm of the soil in the three reference study areas (Fig. 4). An exponential decline of ¹³⁷Cs with depth was found in the profiles, being typical of uncultivated areas (Walling and Quine, 1995; Navas et al., 2005; Gaspar and Navas, 2013).

The retention of ¹³⁷Cs at the first portion of the profile could be attributed to organic matter, since this soil property was highly correlated with ¹³⁷Cs mass activity in the different reference sites. The key role of organic matter in fixing the radionuclide has been shown elsewhere (e.g. Kim et al., 2006; Gaspar et al., 2013). Organic matter is typically bound by non-crystalline minerals in volcanic soils, and one of their common characteristics is accumulation of relatively large quantities of organic matter (Warkentin and Maeda, 1980). Both non-crystalline clays and organic matter modify two key soil variables: they increase moisture retention capacity, reducing the amount of water that percolates; and they increase cation-exchange capacity, and thereby, ¹³⁷Cs adsorption sites. A strong retention of ¹³⁷Cs at the soil surface was found in volcanic soils from Iceland, which was attributed to the

presence of both non-crystalline minerals and organic matter (Sigurgeirsson et al., 2005).

The depth of the maximum concentration of ¹³⁷Cs in Trevelin and Los Alerces National Park was 0–5 cm. At the Esquel site, instead, some profiles showed the highest values at 0–5 cm and others at 5–10 cm. This result, together with finding ¹³⁷Cs at greater depth in a soil profile from Esquel, suggest bioturbation processes, since burrow systems by *Ctenomys* sp. are frequent in different natural environments from ecotone of Patagonian Andean region (Tammone et al., 2012). It is known that burrowing animals can affect the ¹³⁷Cs distribution in soils to greater depths (Müller-Lemans and Van Dorp, 1996).

The depth of the maximum concentration found in our study area, agrees with studies done in the northern hemisphere, where ranged between 0 and 12 cm (Gaspar and Navas, 2013; Jagercikova et al., 2015; Quijano et al., 2016a; Abraham et al., 2018).

4.3. ¹³⁷Cs patterns of gain and loss: relationship with land use

Both ¹³⁷Cs mass activity and inventory, significantly differed between the reference soils from Esquel (i.e., pristine, flats and stable

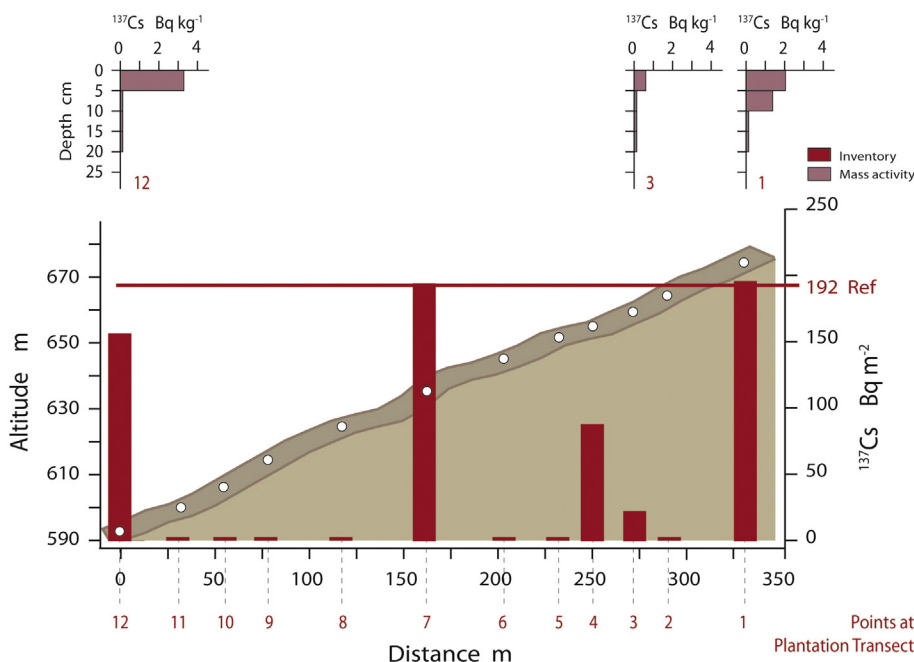


Fig. 7. Topography of the plantation transect, ¹³⁷Cs inventories (Bq m⁻²) for the twelve soil profiles (placed ca. 25 m apart) and detailed information of ¹³⁷Cs mass activity (Bq kg⁻¹) for sectioned soil profiles 1, 3 and 12.

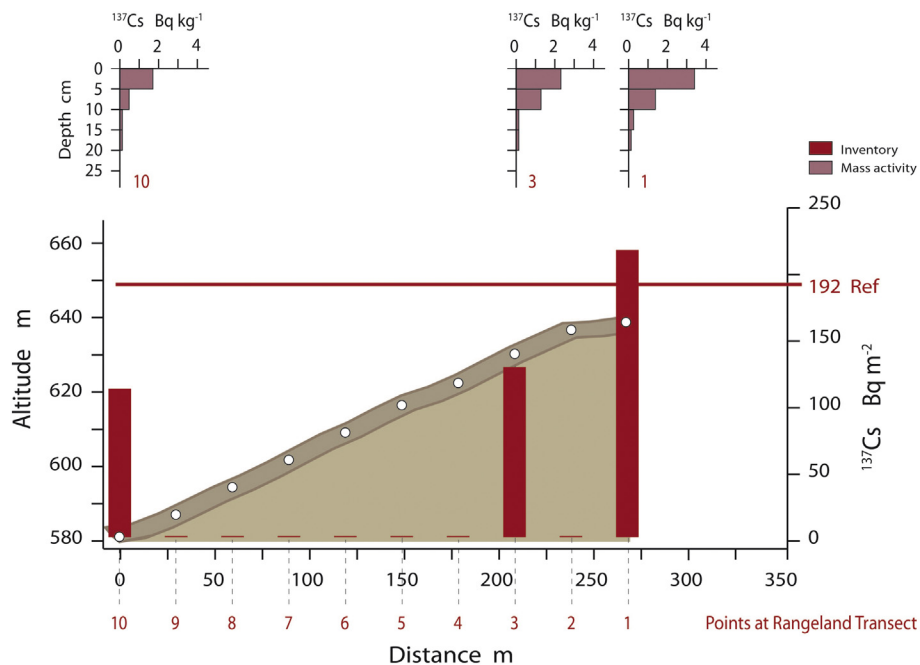


Fig. 8. Topography of the rangeland transect, ^{137}Cs inventories (Bq m^{-2}) for the ten soil profiles (placed ca. 25 m apart) and detailed information of ^{137}Cs mass activity (Bq kg^{-1}) for sectioned soil profiles 1, 3 and 10.

sites) and the different land uses (i.e., forest under grazing, pine plantation and rangeland), where marked ^{137}Cs losses were recorded (Tables 3 and 5).

Soil properties varied according to the land use. Native forest showed the greatest values of pH NaF, organic matter and silt granulometric fraction, and unlike the other land uses, some profiles presented non-crystalline minerals (imogolite) (Table 4). Despite these differences in soil properties, the values of ^{137}Cs , both mass activity and inventory, did not significantly differed between the different land uses (Table 5). Most samples showed ^{137}Cs values lower than the reference value for Esquel, suggesting loss of soil as consequence of erosion processes. As much as 72% (8 out of 11), 83% (10 out of 12) and 90% (9 out of 10) of sample points from native forests, plantation and rangeland, respectively, had ^{137}Cs values lower than the reference sites (Figs. 5, 7 and 8). Besides, 45%, 58% and 70% of sample points from native forests, plantation and rangeland, respectively, showed ^{137}Cs values below the limit of detection.

The ^{137}Cs values in native forest patches under grazing suggest that erosion/sedimentation processes were active in the last 50 years, showing eroding, but also aggrading profiles. Most eroding profiles have lost between 80 and 100% of ^{137}Cs , whereas gain of ^{137}Cs just ranged between 4 and 19% (Fig. 5). The loss of ^{137}Cs was related to the degradation condition of the forest patch, with more critical values in the most degraded Forest B (Fig. 2). It is known that ecological and management factors, like canopy cover, litter cover, and forest vertical stratification, control both soil degradation and erosion (Pennock and Van Kessel, 1997; Morales et al., 2013; Altieri et al., 2018). The aggrading profiles, with highest ^{137}Cs mass activity, also showed the highest porosity and organic matter content. Recent studies developed on native forests under grazing in Patagonia suggest that allophanized soils, even under high livestock stocking rates, keep high physical-chemical fertility, which was explained by resilience capacity of volcanic soils (Gomez et al., 2018). Although our results at Esquel site, on non-allophanized soils, also showed high fertility in native forest under grazing (Table 4), the high loss of ^{137}Cs evidence severe erosion processes.

Along the plantation transect, loss of ^{137}Cs was recorded, except at midslope (P7) and at the highest part of the hillside (P1), where the

^{137}Cs content was similar to the reference value. These results suggest that at P7, a balance between soil loss and gain exists, while uppermost (P1), where slope diminishes and pines coexist with native shrubs, soil appears not to have suffered neither erosion nor soil accumulation (Fig. 7). The ^{137}Cs mass activity, varying along the transect, was highly correlated with organic matter content and porosity. Numerous studies have shown that both, loss/gain ^{137}Cs as well as soil properties, are closely related to topography along the hillsides (e.g. Gaspar et al., 2013; Navas et al., 2017).

Along the rangeland transect, the loss of ^{137}Cs ranged between 33 and 100%, respect to reference value (Fig. 8). Only at the highest part of the hillside (P1), where slope degree diminishes and soil is protected by shrubs and native trees (Fig. 2c), radionuclide gain was recorded, and the ^{137}Cs inventory resulted 13% higher than the reference value (Fig. 8). This result suggests that patches of native shrubs and trees have played a role in reducing erosion in this degraded rangeland.

The vertical distribution of ^{137}Cs under the different land uses was similar as in reference sites, and exhibited the characteristic exponential decline of the radioisotope activity with depth of uncultivated areas (Walling and Quine, 1995). Non eroding profiles showed the highest ^{137}Cs penetration depth (Figs. 6, 7 and 8). The depth of the maximum concentration of ^{137}Cs was 0–5 cm for all the profiles, and ^{137}Cs activity was significantly correlated with organic matter under the different land uses, agreeing with studies worldwide on different soils (Quine et al., 1994; Mabit et al., 2008; Gaspar et al., 2013; Alewell et al., 2014; Quijano et al., 2016b; Mesrar et al., 2017).

4.4. Erosion rates in Esquel: relationship with land use

Rangelands from the transition (ecotone) between the Andean forests and the Patagonian steppe, as those located in Esquel, have suffered the greatest human pressure and overgrazing by livestock and sheep for most than a century (Veblen and Lorenz, 1988). The great loss of ^{137}Cs found in the rangeland, proving soil loss, agrees with the history of use of these ecotonal areas. However, results also suggest that under both, patches of native forest and pine plantation, soil erosion during the last 50 years was high. The patches of native forests appears as isolated patches in an anthropized area under grazing, both currently and in the

past, and erosion indicators can be evidenced in the surroundings, such as bare soils, superficial gravel and tree root exposure (Fig. 2). On the other hand, the pine plantation, with 32 and 21 years old, at bottom and mid-slopes, and at upper slope, respectively, replaced a degraded and shrubby native forest, which had been used for grazing and extracting wood. It is known that pine plantations do not efficiently protect the soil until crowns are quite close and a fresh litter layer is formed (Maestre and Cortina, 2004; Gu et al., 2013), which, in the study area, is reached when the plantations are around 10–13 years old (Tarabini et al., 2019).

Both native forest patches and pine plantation were sampled in microsites where, currently, the soil is completely covered by litter. In fact, both vegetation types showed a continuous litter cover, which protects the soil and minimizes the erosion rates (La Manna et al., 2016). Studies on pine plantations based on ^{137}Cs , found minimum values of soil loss in stand with high canopy cover and biomass (Altieri et al., 2018). Thus, ^{137}Cs results are obviously conditioned not only by current use but also by land use in the last decades.

In each land use, ^{137}Cs greatly varied according to microtopography and soil protection, however, the different land uses agree in the existence of many sample sites where ^{137}Cs values were below the limit of detection. Although the adjustment of equations to determine soil loss rates based on radiometric techniques could require a larger sampling of the reference inventory (Sutherland, 1994; Mabit et al., 2002), and radionuclides that allow determinations in shorter terms should be considered (Gaspar et al., 2017), negligible values of ^{137}Cs suggest the loss of at least 15 cm of soil in the last 50 years in many sample points under the different land uses. This value equals $30 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$. According to bulk density values found under different land uses (Table 4), erosion rates in Esquel reached values of at least $22 \text{ t ha}^{-1} \text{ year}^{-1}$ in native forests under grazing and $33 \text{ t ha}^{-1} \text{ year}^{-1}$ in plantation and rangeland slopes.

Setting tolerable erosion rates requires taking into account reference rates of soil formation, resulting from mineral weathering as well as dust deposition (Verheijen et al., 2009). Recent studies assessed the rates of soil formation in pine plantations from Esquel, by entrapping particles, finding mean rates of 0.1 mm year^{-1} or $1 \text{ m}^3 \text{ ha}^{-1} \text{ year}^{-1}$, equivalent to $0.9 \text{ t ha}^{-1} \text{ year}^{-1}$ (Tarabini et al., 2019). These soil formation rates, similar to those assessed in temperate regions of Europe (Verheijen et al., 2009) highly differed from the erosion rates estimated in our study. These results highlight the requirement of reliable quantification of soil erosion rates in Patagonia and the urgent need of applying effective soil conservation measures.

5. Conclusions

This work is the first approach to the use of radioisotopes in relation to erosion in the Andean Patagonia region of Argentina. The ^{137}Cs reference inventories varied according to the edaphoclimatic gradient. Allophanized soils showed the highest ^{137}Cs values, even higher than the expected according to the precipitation amount. This result suggests a high adsorption of ^{137}Cs by allophanized soils, related to the non-crystalline nature of the clays and to the high content of organic matter.

In the subhumid study area, high erosion rates were found under different land uses. These results emphasize the high erodability of non-allophanized volcanic soils, which seem to suffer severe loss of soil under any land use, once the original vegetation and soil cover are lost.

Reference inventory values and the loss of the radionuclide in sites with high anthropic intervention, show the potential for using ^{137}Cs measurements for assessing erosion processes in these latitudes

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