Mapping the spatial distribution of global ^{137}Cs fallout in soils of South America as a baseline for

Earth Science studies

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Abstract

Owing to the rapid expansion of agriculture in South America in recent decades, soil erosion and fine sediment supply to river networks, which lead to deleterious on-site and off-site environmental impacts, are exacerbated in intensively cultivated catchments. Measuring soil inventories of bomb-derived fallout radiocesium (^{137}Cs) bound to fine particles is one of the few techniques available to reconstruct soil redistribution rates and evaluate the sustainability of farming practices over the recent phase of agricultural intensification (1960s–2020). However, information about the spatial distribution of ^{137}Cs fallout across the soils of South America remains scarce, and the published data has not been synthesized at the scale of this subcontinent so far. The objective of the current research is therefore to quantify and map the initial ^{137}Cs fallout at the scale of South America, based on the compilation of published ^{137}Cs inventories, additional measurements conducted on undisturbed soil profiles and digital soil mapping as this baseline information may be useful for a wide range of Earth Science applications. A database of ^{137}Cs inventories at 96 reference sites (i.e. areas without soil erosion nor accumulation) has been compiled for a variety of soil profiles (Argentina = 10, Brazil = 34, Chile = 46, Uruguay = 5, French Guiana = 1) located between 5.3° North latitude and 53° South latitude. The spatial distribution of ^{137}Cs fallout was shown to be highly latitude-dependent, with a maximum in the 30–50° South latitude band. There were higher fallout levels than expected between 20 to 60° South latitude compared to the previous estimations made by UNSCEAR. A partial least square regression approach based on rainfall data and geographical information as covariates was used to create a baseline map of ^{137}Cs fallout in soils of continental South America. This baseline map provides a powerful reference dataset to anticipate the order of magnitude of ^{137}Cs inventories in undisturbed soil profiles collected in Brazil and Southern Chile and for numerous other applications in Earth Sciences. The potential application of the ^{137}Cs inventory technique in countries of South America in general, and in regions with vulnerable ecosystems threatened by the expansion of agricultural activities in particular, is discussed in light of this comprehensive literature review. Furthermore, the regions (i.e. the North-Western part of the continent) where additional samples should be collected in priority to improve this baseline map are outlined. Our results demonstrate that ^{137}Cs inventories are sufficiently high to

investigate soil redistribution rates in most of South American countries where detectable levels of ^{137}Cs can be expected to be found in sites exposed to erosion.

Keywords: Digital soil mapping, Caesium-137, Soil erosion, Soil redistribution rates, Reference soil sites

1. Introduction

Soil is essentially a non-renewable resource over the human timescale (Lal, 2015). The future use of this valuable resource is threatened by anthropogenic activities such as the intensification of agricultural practices (Keesstra et al., 2016). The Great Acceleration period that started during the second half of the 20th century induced a significant increase of soil erosion and degradation (Steffen et al., 2015), leading to extensive on-site and off-site impacts. On-site, soil erosion threatens soil fertility and agricultural yields (Bakker et al., 2007; Bakker et al., 2004; Vanwalleghem et al., 2017). Fine-grained sediment transported off-site, which may be associated with nutrients and contaminants, leads to the reduction of water quality, the degradation of aquatic habitats and reservoir siltation (Becker et al., 2009; Evrard et al., 2007; Foucher et al., 2014). Fine sediment supply induced by soil erosion processes is exacerbated in intensively cultivated catchments, leading to deleterious consequences for river systems (Owens et al., 2005) and to the disturbance of global biogeochemical cycles (Quinton et al., 2010).

In order to evaluate the sustainability of agricultural practices and promote effective soil management practices, a fundamental prerequisite is to quantify soil erosion rates. Measuring soil inventories of fallout radionuclide Caesium-137 (^{137}Cs , $t_{1/2} = 30.2$ years) has shown great potential to provide retrospective information on soil redistribution rates over the medium-term (Ritchie and Ritchie, 2007), despite the critiques made on this method (Parsons and Foster, 2011). The latter authors recommended that future studies should present their results in light of the underlying hypotheses (e.g. the spatial uniformity of fallout at the local scale). Despite the ongoing debate in the literature,

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77 this technique has been increasingly adopted as a field-based approach to quantify soil erosion or
78 deposition since the 1980s in several catchments around the world (Chartin et al., 2013; Fukuyama et
79 al., 2005; Mabit et al., 2018; Mabit et al., 2008; Navas et al., 2005; Zapata, 2002).

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80 ^{137}Cs is an artificial radionuclide generated as a product of the atmospheric nuclear weapon tests
81 carried out between 1945 and 1980, or released by nuclear accidents (e.g. Chernobyl in Ukraine,
82 Fukushima in Japan) (Steinhauser et al., 2014). The introduction of ^{137}Cs into the global environment
83 also coincides with the transformation of farming practices initiated in the 1960s through the
84 increasing use of external inputs and heavy machinery (Camargo et al., 2017). When deposited onto
85 the soil surface, ^{137}Cs is rapidly and almost irreversibly bound to fine particles, as its desorption by the
86 natural chemical processes infrequently occurs in continental environments (Tamura, 1961; Tamura
87 and Jacobs, 1960). Assessment of erosion and deposition rates is commonly based on the comparison
88 of ^{137}Cs inventories (Bq m^{-2}) in undisturbed soil profiles with those inventories measured at locations
89 affected by soil redistribution in the landscape (Loughran et al., 2002; Walling and Quine, 1990). A
90 reference site is defined as an undisturbed soil profile where neither erosion nor deposition has
91 occurred since the period of radioactive fallout. Accordingly, it contains the cumulative atmospheric
92 fallout input at the site reduced by radioactive decay only (Zapata, 2003). Measuring ^{137}Cs inventories
93 in reference sites may therefore provide the only indirect approach available nowadays to reconstruct
94 the initial fallout released by the nuclear weapon tests on these soils.

95 Understanding the spatial distribution of initial ^{137}Cs fallout is important for several scientific
96 disciplines, including medical science (Gilbert et al., 2002; McCarthy, 1997; Simon et al., 2006) as well
97 as various fields in Earth Science, with applications in ocean, atmosphere and soil-related studies
98 (Buesseler and Benitez, 1994; Ehhalt, 1973; Jagercikova et al., 2017). The first estimation of the fallout
99 spatial pattern was made by the United Nations Scientific Committee of the Effects of Atomic Radiation
100 (UNSCEAR, 1962) in order to evaluate the health hazard of fallout radionuclides. Based on the results
101 of a long-term monitoring programme of global fallout deposition, a uniform distribution model for

10-degree latitudinal bands was established in the UNSCEAR report published in 2000. In parallel, soil erosion and soil-to-crop transfer studies have provided more accurate local information on the spatial distribution of initial ^{137}Cs fallout worldwide (Owens and Walling, 1996; Ritchie and McHenry, 1990; Walling and He, 2000). The deposition of fallout radionuclides onto the Earth surface is conditioned by two major factors: latitude and rainfall. Ground deposition of radionuclides from the global fallout was shown to be highly latitude-dependent, with a maximum in the 40-50° bands in both hemispheres (UNSCEAR, 2000). In addition, the activity of ^{137}Cs deposited on the ground and its variation from one location to another is closely linked to the annual rainfall rates, as a consequence of precipitation scavenging (Bouisset et al., 2018; La Manna et al., 2019; Le Roux et al., 2010; Malakhov and Pudovkina, 1970). Moreover, relationships between monthly rainfall rates and ^{90}Sr fallout were observed in South America, with a peak during the months of June-August (Volchok, 1965; Volchok, 1966). Based on these two factors, digital mapping using geostatistical approaches has been conducted at different scales to map bomb-derived ^{137}Cs fallout (Almgren et al., 2006; Aoyama et al., 2006; Chappell et al., 2011a; Furuichi and Wasson, 2013; Meusburger et al., 2020; Palsson et al., 2006). Although reconstructions of baseline ^{137}Cs inventories were made for Europe, Australia and Eastern Asia, information about the spatial distribution of ^{137}Cs in soils of South America remains scarce, which requires further investigation to determine the potential for application of ^{137}Cs for Earth Science studies in this subcontinent. Of note, the ^{137}Cs inventories in soils of the Southern hemisphere are usually sufficient to be measurable by gamma spectrometry when using appropriate low-background detectors during sufficient counting times (FAO/IAEA, 2017).

Quantifying ^{137}Cs inventories in soils and calculating the corresponding soil redistribution rates during the intensive cultivation period (1960 to present) is particularly important to evaluate the sustainability of agricultural practices implemented in South America during the last several decades to maintain erosion at sustainable levels (Minella et al., 2014). During the last 50 years, most countries of the subcontinent have abandoned conventional tillage to implement no-till farming (Montgomery, 2007). This change in practice has been accompanied by an increase in the size of cultivated areas and their

overall productivity (Wingeyer et al., 2015). However, no-till farming has been widely implemented as a single conservation measure without additional practices to reduce soil loss, such as crop rotation or contour cropping and terracing (Didone et al., 2019; Montgomery, 2007). As a consequence, soil erosion by water remains the main soil degradation process in agricultural land of South America and its real magnitude remains debated among agricultural and scientific communities (FAO, 2015).

To provide independent estimations of soil redistribution rates, appropriate reference soil sites are increasingly difficult to find in these intensive agricultural areas, given the growing extent of crops on almost all available land areas. The indirect estimation of ^{137}Cs inventories in reference soil sites of South America is therefore required to assess soil redistribution rates with the ^{137}Cs technique. Based on the compilation of published ^{137}Cs inventories and additional measurements conducted on undisturbed soil profiles, the primary goals of this study are (1) to present and discuss the spatial distribution of ^{137}Cs inventories in reference soil sites across South America, (2) to map the initial ^{137}Cs fallout at the scale of South America using a digital mapping approach in order to provide estimates at unsampled reference locations, and (3) discuss the potential for application of this technique and other Earth Science approaches in countries of South America. Although this new map cannot replace the sampling of profiles in local reference areas, it will provide useful information for guiding and designing future Earth Science studies in South America using ^{137}Cs global fallout.

2. Spatial distribution of bomb-derived ^{137}Cs in soils of South America

2.1. Global fallout following nuclear weapon tests

The UNSCEAR reports provide the main source of information regarding ^{137}Cs deposition worldwide. Early studies about the production and transport of nuclear weapon debris started in 1962 (UNSCEAR, 1962). It is currently accepted that a total of 502 atmospheric tests, with a total fission and fusion yield

of 440 Mt, were conducted from the mid-1950s to 1980 (UNSCEAR, 2008). Following the extensive tests of atmospheric nuclear devices in 1962, the deposition of ^{137}Cs reached a peak in 1963 and 1965 in the Northern and Southern hemispheres, respectively (Cambray et al., 1989; Turney et al., 2018). The signing of the Partial Test-Ban Treaty on August 5, 1963 by the United Kingdom, the Soviet Union and the United States has contributed to the reduction of global emissions of artificial radionuclides into the environment. In soils of South America, no trace of Chernobyl and Fukushima Daichii-derived fallout radionuclides were detected, which makes the atmospheric weapon test fallout the single significant source of artificial radioactivity in this part of the world (Steinhauser et al., 2014).

The radioactive debris emitted as a result of atmospheric nuclear tests were distributed between the surface of the ground or water and the tropospheric and stratospheric regions, depending on the type of test (offshore barge, top of a tower, under a tethered balloon, etc.), the location (altitude and firing latitude) and the power (kilotonnic or megatonnic). The majority of the radioactive debris was dispersed into the stratosphere (referred to as “stratospheric fallout” or “global” fallout). The activity that did not reach the stratosphere is referred to as “local / regional” fallout and “tropospheric fallout” (UNSCEAR, 2000).

Local fallout includes radioactive aerosol particles generally larger than 50 μm in size that are deposited within a radius of about 100 km from the epicentre of the explosion (Garcia Agudo, 1998). Tropospheric fallout is characterized by smaller aerosols that were deposited with a mean atmospheric residence time of up to 30 days. During this period, the debris were dispersed within the latitude band of the initial injection and followed wind-driven trajectories, before falling to the ground as a consequence of precipitation scavenging (Bennett, 2002). Stratospheric fallout, which makes up a large portion of total deposition, consists of particles that are transported into the stratosphere, dispersed and then deposited globally, most of which occurs in the hemisphere of the initial injection. Stratospheric deposition accounts for the majority of global long-lived fission product residues (UNSCEAR, 2000).

In order to estimate radionuclide deposition following nuclear detonations, two long-term monitoring programmes were established during the nuclear weapon test period. The first programme, conducted by the United Kingdom Atomic Energy Authority (UKAEA) consisted of 8 stations located in the United Kingdom and 18 stations installed in the rest of the world. The second programme, conducted by the Environmental Measurements Laboratory in the United States (EML), as early as in 1954, consisted of 177 stations distributed worldwide. This monitoring programme, based on monthly ^{90}Sr fallout sampling, was the largest and the most widely distributed and therefore, it has been adopted by the UNSCEAR to estimate the total hemispheric annual deposition of ^{90}Sr (UNSCEAR, 1993). The global distribution of ^{137}Cs was assessed assuming a $^{137}\text{Cs}/^{90}\text{Sr}$ fission yield ratio of 1.5, and a uniform distribution model for each 10-degree latitude band was published in the UNSCEAR (2000) report.

The latitudinal distribution of ^{137}Cs described by UNSCEAR should be taken with caution as 50% of the data is missing from this monitoring programme (Evrard et al., 2020). Indeed, a significant proportion of data is lacking for monitoring stations ($n=34$) located in South America. In all, 59% of monthly ^{90}Sr fallout data is missing between 1954-1976, as most of the monitoring stations did not have continuous records (Health and Safety Laboratory, 1977). In addition, when studies investigating ^{137}Cs inventories in undisturbed soil profiles are conducted at regional or continental scales, the proportion of global fallout estimated by UNSCEAR appears to be underestimated (Aoyama et al., 2006; Chappell et al., 2011a; Schuller et al., 2002; Tagami et al., 2019). Regarding the information currently available for ^{137}Cs fallout at the scale of South America, the only reference is a study published by Garcia Agudo (1998) who also used results of these monitoring programmes to map the global inventory of ^{137}Cs from nuclear tests worldwide. Given the wide range of applications of ^{137}Cs in the scientific literature, as well as the debate raised on its use as a tracer for soil erosion assessment (Mabit et al., 2013; Parsons and Foster, 2011; Parsons and Foster, 2013), a more accurate characterization of the initial fallout of bomb-derived ^{137}Cs and its spatial distribution across South America is fundamental to provide a baseline for future studies.

2.2. Data collection on ^{137}Cs in reference soil sites of South America

A literature survey was carried out using Web of Science databases on March 1, 2020. Most data were found in peer-reviewed scientific articles published in English and Portuguese languages (n=17), although inventories were also taken from unpublished PhD manuscripts (n=2). The search keywords 'soil erosion', 'cesium-137', ' ^{137}Cs ' and 'inventorie(s)' were used in isolation and/or combination with additional keywords containing the country names of South America. In addition, six scientific articles that were not identified during the initial Web of Science search were included in the literature compilation during the review process. Three conditions were necessary to accept data: (1) the data were provided in Bq m^{-2} , (2) sampling locations were provided or easy to obtain and (3) information about sampling procedures was detailed. The sampling year or the year of decay-correction were also required to standardise all the data compiled. Seven studies did not mention the sampling year or the year of decay-correction. In this case, we made the assumption that sampling was conducted 4 years before the report publication, corresponding to the mean duration between sampling and publication in the current literature survey. All the data was decay-corrected to 2020 according to Eq. (1)

$$^{137}\text{Cs}, 2020 (\text{Bq m}^{-2}) = ^{137}\text{Cs}_{\text{literature}} (\text{Bq m}^{-2}) e^{(-\lambda * t)} \quad (1)$$

where λ is the decay constant of ^{137}Cs ($\lambda = \ln 2 / 30.2 \text{ y}$) and t is the time (year) since the sampling year as reported in the corresponding article or estimated following the above-mentioned method. Each entry in the database corresponds to one undisturbed soil profile for which the ^{137}Cs inventory has been calculated, its geographical coordinates (latitude, longitude, elevation), annual rainfall rates, sampling method, ^{137}Cs inventory and the associated standard deviation estimated by the author and the associated decay-corrected values calculated from Eq. (1). In total, a database of 103 ^{137}Cs inventories at reference soil sites has been compiled, in four different countries and one overseas

department of France (Antarctica = 2, Argentina = 10, Brazil = 38, Chile = 50, Uruguay = 2, French Guiana = 1) between 5.3° North latitude and 62° South latitude and from 109.3° to 34.9° West longitude (Fig. 1). Mean annual precipitation, the year of decay-correction, altitude and standard deviation were reported for 98, 82, 71 and 40 ¹³⁷Cs inventories calculated at these reference soil sites, respectively. Four ¹³⁷Cs inventories from Easter Island (Pacific Ocean) and Antarctica were removed as they were not obtained on the main South-American continent. Of note, four inventories obtained near the Amazon River (0 and 1 Bq m⁻² respectively) and in Southern Brazil (5 and 90 Bq m⁻²), analysed by Handl et al. (2008) may have been sampled in erosional areas rather than at reference sites, which may have an impact on the results of the current research. Conversely, the highest ¹³⁷Cs inventories were found in the central part of Chile (3113 and 2860 Bq m⁻²), at 40° South latitude (Schuller et al., 2002). These inventories, with elevated ¹³⁷Cs concentrations, may have been sampled in accumulation areas instead of reference locations. These six samples were considered as anomalous.

To gain insight into the accuracy of the baseline map of ¹³⁷Cs fallout derived from this dataset, additional unpublished ¹³⁷Cs inventories in undisturbed soil profiles were used in the present study. In total, 24 soil cores were collected by co-authors in Uruguay down to a soil depth of 20cm to characterize three reference sites. All soil samples were collected using an Eijkelkamp soil sampler (5 cm diameter) and then stored in polyethylene bags. Samples were ground and passed through a 2 mm sieve, then placed in 250 mL plastic Marinelli beakers. Gamma-ray measurements of ¹³⁷Cs (662 keV) were obtained by high purity germanium detector (HPGe) (Canberra) at Laboratorio de Radioquímica (Centro de Investigaciones Nucleares, Universidad de la República, Montevideo, Uruguay) and at Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). In summary, among the 103 ¹³⁷Cs inventories compiled in the literature and the additional three ¹³⁷Cs inventories collected by the co-authors, 96 ¹³⁷Cs inventories (Table 1) were used to perform the statistical analysis presented below.

2.3. Spatial distribution of ^{137}Cs inventories in South America

According to the current literature review, the highest ^{137}Cs inventories in reference soil sites were found in the latitude band between 40 to 50° South (Fig. 2). From this location, inventories decreased towards both the Equator and Southern Patagonia, where the mean ^{137}Cs inventories were 97 and 279 Bq m⁻², respectively. The distribution of ^{137}Cs inventories in reference soil sites of South America is highly latitude-dependent, with a maximum in the 30-50° South latitude band, which is slightly higher than that proposed by UNSCEAR for that same latitude range. In contrast, the ^{137}Cs inventories in the 0-20° South latitude band found in the literature is slightly lower than that proposed by UNSCEAR (Fig. 3). Attention should be paid as published inventories were not found in the literature for the North-Western part of South America. Accordingly, the distribution determined from ^{137}Cs inventories in undisturbed soil profiles is mostly valid for Brazil, Uruguay, Argentina and Chile.

The typical pattern of ^{137}Cs deposition with latitude can be explained by the preferential exchange of air between the stratosphere and troposphere at mid-latitudes, as well as the air circulation patterns in the troposphere that both lead to an increased deposition of fallout radionuclides in the temperate regions and to a decreased deposition in the equatorial and polar regions (UNSCEAR, 2000). Indeed, the upward circulation that occurs in the equatorial region, known as Hadley circulation, is characterised by air masses rising from the ground into higher regions of the troposphere (Martin and McBride, 2012). According to UNSCEAR, this particular circulation pattern may explain the lower ^{137}Cs inventories in reference soil sites analysed in this latitude band, despite high annual rainfall rates reported in the Amazonian region (Handl et al., 2008). In contrast, ^{137}Cs inventories in reference soil sites are higher despite lower annual rainfall rates between 30 to 50° South latitude as a consequence of the Ferrel circulation, where air masses fall back down in the vicinity of the mid-latitudes, leading to an increase of radionuclide deposition in this part of South America (UNSCEAR, 2000).

A significant linear relationship was found between ^{137}Cs inventories in reference soil sites and annual rainfall rates provided by the authors ($R^2=0.43$, $n=92$, $p < 0.001$). However, a wide dispersion of ^{137}Cs inventories values is found for similar levels of precipitation (Fig. 4). In contrast, when reorganising the data according to the climatic circulation cells in which they are located, a significant relationship is observed between mean annual rainfall rates and ^{137}Cs inventories in soils of Argentina and Chile ($R^2=0.72$, $n=54$, $p < 0.001$). Furthermore, another linear relationship, although less significant, is found for those reference soil sites located between the Equator and mid-latitudes ($R^2=0.39$, $n=38$, $p < 0.001$). Contrary to what has been observed for precipitation, no significant relationship was found between the altitude reported by the authors at the reference sites and the ^{137}Cs inventories ($n=74$). However, when the analysis is performed for ^{137}Cs inventories in reference soil sites located at the same latitude although at a different altitude, it can be observed that ^{137}Cs inventories tends to increase with the altitude (data not shown). This observation confirms that made by Handl et al. (2008) who stated that maximum values were observed in regions of high altitude between 23° and 29° South latitude. In Chile, Schuller et al. (2002) found a similar relationship for reference soil sites located in the central part of the country, with higher values in elevated altitude areas, which are also exposed to higher rainfall. Furthermore, a low correlation is observed between the altitude of the sampling sites and the ^{137}Cs inventories in reference sites located between the mid-latitudes and Southern Chile ($R^2 = 0.20$, $n=52$, $p < 0.01$). A step wise multiple regression indicated a significant relationship between observed and predicted ^{137}Cs inventories with a model including geographical information and annual rainfall rates ($R^2 = 0.74$, $n = 71$, $p < 0.001$) (Fig. 5).

3. Mapping ^{137}Cs initial fallout in South American soils

3.1. Context

To the best of our knowledge, only very few studies (n= 7) were conducted to map bomb-derived ^{137}Cs fallout (Table 2). The spatial distribution of ^{137}Cs deposition was assessed when results of global fallout long-term monitoring programmes were available during the nuclear weapon tests period. By means of a GIS-based approach, Wright et al. (1999) used ^{137}Cs deposition data from the Arctic Monitoring and Assessment Programme (AMAP) with annual rainfall rates for the period 1955-1985 to predict ^{137}Cs deposition at the scale of the Arctic. Similarly, Pálsson et al. (2006) used activity concentrations of global fallout in precipitation at Rjúpnahæð, in addition to ^{137}Cs measurements in undisturbed soil profiles, to predict the spatial variation in global fallout of ^{137}Cs in Iceland. In Australia, Chappell et al. (2011a) used indicator co-simulation between 141 inventories in reference soil sites and gridded mean annual rainfall (1954–1990) to create a baseline map of ^{137}Cs fallout for Australian soils. More recently, Meusburger et al. (2020) used a digital soil mapping approach (McBratney et al., 2003) to predict the proportions and sources of artificial radionuclides in soils of several countries of Western Europe (France, Belgium, Switzerland, Southern Germany and Northern Italy). Their approach used a generalized additive models (GAM) with environmental factors.

3.2. Digital mapping of ^{137}Cs fallout in soils of South America

Mapping in the current research is based on a digital soil mapping (DSM) approach where field observations are combined with environmental data (covariates) and a statistical model to map the ^{137}Cs inventories at the scale of South America. Once a model is fitted to the data, it can be used to spatially predict the soil attribute at unobserved locations based on the observed environmental data at these locations. Environmental data should represent influential factors that explain the spatial variation of the target soil attribute. As previously mentioned, ^{137}Cs inventories in reference soil sites are strongly correlated with rainfall and latitude. We therefore retained two types of covariates : a spatially interpolated monthly rainfall database (WorldClim, <https://www.worldclim.org/>) at a

resolution of 30-arc seconds, calculated from the 1950-2000 period (Hijmans et al., 2005), and the spatial coordinates (X and Y in meters).

Based on a cross validation procedure, a partial least square regression (PLSR) approach was selected.

The theory underlying PLSR has been described in several statistical textbooks and articles (Höskuldsson, 1988; Tenenhaus, 1998). PLSR could be deemed as a generalization of the multiple linear regression (Gerlach et al., 1979). PLSR is of particular interest because, unlike multiple linear regression, it can analyze noisy data with numerous collinear variables.

The statistical analyses as well as the DSM procedures described in this section were carried out with the R software (Team R Core, 2013) and the following packages: caret (Kuhn, 2008), sf (Pebesma, 2018), ithir (Malone, 2015), ggplot2 (Wickham, 2016), raster (Hijmans et al., 2015), corrplot (Wei et al., 2017) and clhs (Roudier, 2011).

Figure 6 shows the correlogram plot and the associated coefficients of correlation. The highest correlation was found between ^{137}Cs inventories and average monthly rainfall in August and September ($r=0.7$, $p < 0.001$), followed by average monthly rainfall in May, June, July and average annual rainfall ($r=0.6$, $p < 0.001$), longitude ($r=0.4$, $p < 0.001$) and latitude ($r=0.3$, $p < 0.01$). Of note, no correlation was observed for elevation. Based on this preliminary statistical analysis, seven covariates (latitude, longitude, average monthly rainfall in June, July, August, September and mean annual rainfall rates) were selected to perform the PLSR.

Following the cross validation procedure, we showed that our quantitative model explained 46% of the ^{137}Cs variability observed and tended to underestimate inventories $> 800 \text{ Bq m}^{-2}$. The predicted ^{137}Cs inventories followed a right-skewed statistical distribution, with an average of 348 Bq m^{-2} and a standard deviation of 237 Bq m^{-2} . Highest values (between 1501 to 2057 Bq m^{-2}) were found in Colombia and few areas of the Andean Cordillera, while the vast majority of ^{137}Cs inventories lower than 100 Bq m^{-2} were found in the North-Eastern part of Brazil (Fig. 7). Surprisingly, high ^{137}Cs

inventories are predicted for countries located near the equator, where fallout should be the lowest according to UNSCEAR (2000). Of note, the highest inventories are mainly located along the Andean Cordillera at high altitudes. Taking into account that the model does not include elevation as a covariable, the deposition pattern of ^{137}Cs is closely linked to that of rainfall rates and it may reflect the orographic effects that occur in mountainous environments. However, these mountainous areas predominantly consist of rock outcrops and bare soil surfaces. Consequently, areas above 1,800 m a.s.l have been masked using the GMTED2010 30-arc-second elevation database (Danielson and Gesch, 2011). As previously mentioned, most of the ^{137}Cs inventories in undisturbed soil profiles used to develop the digital soil mapping approach were measured in the eastern part of South America and in Southern Chile. The estimates provided for the other regions (i.e. northern and western parts of the subcontinent) should therefore be interpreted with caution. Additional sampling in these areas will be necessary to improve model predictions of ^{137}Cs global fallout in South America.

The baseline map of ^{137}Cs inventories in reference soil sites (Bq m^{-2} , 2020) with a spatial resolution of 2 km (projection: WGS 84 - World Geodetic System 1984) was compiled using ESRI ArcGIS 10.6 Desktop. The validity of the proposed approach to predict the spatial distribution of the reference levels of fallout ^{137}Cs at the subcontinental-scale of South America relies on the assumption that studies listed in our database followed the recommendations to select and sample undisturbed soil sites. Since the coefficient of variation of multiple samples collected to characterize one reference site is not always reported by authors, the observations uncertainties are difficult to quantify. As reported in the literature, ^{137}Cs inventories in reference soil sites have a coefficient of variation of approximately 20%, as a consequence of random and systematic spatial variability, sampling variability and measurement precisions (Loughran et al., 2002; Owens and Walling, 1996; Pennock, 2000). Accordingly, the map predictions provided in this study for each grid should be interpreted as a trend rather than a single accurate value.

A non-parametric bootstrap approach was used to quantify the prediction uncertainties of the map (Efron and Tibshirani, 1994; Liddicoat et al., 2015; Rossel et al., 2015). In this approach, data used for model calibration was selected using random sampling with replacement, with sample size equal to 95 % of the number of data in the available dataset. This calibration step is iterated 100 times leading to 100 contributing predictions at each prediction location. These predictions collectively constituted an empirical probability distribution (EDP) of the ^{137}Cs inventories. We computed the 95 % prediction confidence interval by subtracting the 97.5% quantile to the 2.5 % quantile of the EDP. For clarity, the coefficient of variation (in %) was computed by dividing the average prediction by the 95% confidence interval (Fig. 8). For most part of South America, the relative error was comprised between 0 and 10 %. The highest uncertainties could be observed in two areas located in the North-Eastern part of Brazil and along the Pacific coast of Peru, where the map predictions of ^{137}Cs inventories in reference soil sites are the lowest (0 to 100 Bq m⁻² in 2020).

3.3. Improving spatial predictions through additional sampling

Sampling design plays an essential role in a digital soil mapping approach as it controls the estimation of the statistical model parameters and also the spatial predictions. A sound sampling design is expected to provide a scheme of representative samples covering the study area with a relatively small sample size for financial and logistical reasons. If it is assumed that the soil property is linked to environmental covariates, a robust strategy is to ensure that the measurements are also uniformly spread in the feature (i.e. covariates) space. However, the collected data of the present study did not follow any sampling design as they were compiled from various independent studies, leading to large areas devoid of sampling observations or places with clusters. Accordingly, an experiment was conducted to generate new sampling locations in order to fill the gap in feature space of the selected environmental variables. This can be achieved using conditioned Latin Hypercube sampling (cLHS)

(Minasny and McBratney, 2006; Wadoux et al., 2019), which guarantees the full coverage of a multivariate feature space. We ran the algorithm with the selected covariates of our model to produce a set of 10 new sampling locations that should be analysed in priority in the future to improve the map in addition to the existing samples (Table 3) (Fig. 7).

4. Potential for application of the ^{137}Cs technique in South America

In South America, where the input of bomb-derived ^{137}Cs fallout has stopped since the 1980s, ^{137}Cs inventories in soils are continuously decreasing as a consequence of radioactive decay. One major challenge is to clearly identify areas where the ^{137}Cs technique could be applied to address Earth Science research questions, or those where it will either become difficult or impossible. Of note, sites exposed to erosion in intensively cultivated catchments could be depleted in ^{137}Cs which would therefore prevent the application of the ^{137}Cs technique in these locations.

Chile is the country of South America where information about ^{137}Cs inventories in reference soil sites is the most documented ($n=48$), including two sites located on Easter Island in the Pacific Ocean). In the central part of Chile, extending from 36 to 42° South latitude, a mean ^{137}Cs inventory of 563 Bq m^{-2} can be calculated from 30 reference soil sites. The high content of ^{137}Cs in soils allowed for the use of the ^{137}Cs technique to quantify soil redistribution rates under different land uses and management practices (Schuller et al., 2003b). Of note, the standard deviation (SD) expressed in Bq m^{-2} was mentioned for 14 reference soil sites (Banfield et al., 2018; Schuller et al., 2003a), while the SD was expressed in Bq kg^{-1} without any information regarding the bulk density of soils sampled for calculating the other ^{137}Cs inventories collected in the country (Schuller et al., 2002). Although ^{137}Cs inventories are lower in the Patagonian part of Chile with a mean value of 280 Bq m^{-2} ($n=16$), the applicability of the ^{137}Cs technique could be explored to investigate the soil response to environmental changes under colder climates (Navas et al., 2018).

In Argentina, ^{137}Cs soil profiles have been collected in the Pampa Ondulada region of Buenos Aires Province (Bujan et al., 2003), in soils of La Plata region (Montes et al., 2013), in natural and semi-natural grassland areas of San Luis Province (Ayub et al., 2008) and in the Patagonian Andean forests (La Manna et al., 2019). These studies showed that ^{137}Cs inventories were sufficient in these regions to calculate soil redistribution rates based on this technique. Of note, there is a lack of ^{137}Cs inventory data in the literature for the Argentinian provinces located between 25 to 40° South latitudes, where inventories in reference soil sites should be the highest of South America according to UNSCEAR (2000). Thus, the ^{137}Cs technique should be successfully implemented in this country exposed to elevated levels of both water and wind erosion (Ares et al., 2016; Mendez and Buschiazzi, 2010).

In Uruguay, only two ^{137}Cs inventory had been documented so far in the literature (Alonso et al., 2012; Tassano et al., 2020). Alonso et al. (2012) assessed soil erosion rates in a forested micro-catchment occupied by eucalyptus plantations within the Río Negro River basin. The three additional ^{137}Cs inventories collected in Uruguay by co-authors of the current research remained in the same order of magnitude, with an average value of $336 \pm 13 \text{ Bq m}^{-2}$. These values are consistent with ^{137}Cs inventories found in the neighbouring Rio Grande do Sul state, in southernmost Brazil, where ^{137}Cs inventories followed an increasing gradient from the Uruguayan border in the South ($315 \pm 22 \text{ Bq m}^{-2}$) to the North ($1022 \pm 292 \text{ Bq m}^{-2}$) (Didone et al., 2019; Handl et al., 2008; Minella et al., 2014).

In general, ^{137}Cs inventories measured in sites of Brazil located below 20°S are high enough to reconstruct soil redistribution rates (Bacchi et al., 2000; Correchel et al., 2005; Didone et al., 2019; Macêdo, 2009; Minella et al., 2014). In addition to these studies where the ^{137}Cs technique has been used successfully, many regions in South America should have received enough fallout to reconstruct soil redistribution rates (Fig 9.A). This includes most of the agricultural regions located in Paraguay and Argentina between 20 to 45°S, as well as Mato Grosso state (Brazil) where intensification in agriculture-forest frontiers is observed (Garrett et al., 2018) (Fig 9. B, C).

Despite the high mean annual rainfall rates observed in the equatorial regions, low ^{137}Cs inventories in reference soil sites were observed in the range of latitudes comprised between 2°N and 10°S. As previously mentioned, one reason that may explain this observation is the upward circulation of air masses as a consequence of Hadley circulation in this latitude band. Soil erosion studies based on ^{137}Cs inventories should be difficult to conduct in the North-Eastern part of Brazil. Furthermore, to the best of our knowledge, no studies on ^{137}Cs inventories in reference soil sites have been conducted in Suriname, Guyana, Venezuela, Colombia and Peru.

One study investigating soil erosion in a mountainous watershed of Ecuador (2°S) with the ^{137}Cs technique was not retained in the current review because geographical information of ^{137}Cs inventories in reference soil site was erroneous (Henry et al., 2013). In this study, no flat undisturbed sites that can serve as typical reference sites were found, and reference sites were selected based on ^{137}Cs activity patterns with depth and land use history. A mean ^{137}Cs reference value of $2260 \pm 330 \text{ Bq m}^{-2}$ was established with no decay-correction date presented. Although this value should be taken with caution, it appears valuable to investigate ^{137}Cs initial fallout in equatorial regions of South America. Indeed, anomalous high values of ^{137}Cs activities were also observed in surface soils of Venezuela (LaBrecque et al., 2001) and one ^{137}Cs inventory in a reference soil site of French Guiana (de Tombeur et al., 2020) was found to be twice than what was expected from the UNSCEAR predictions for this latitude band (1022 ± 293 compared to $509 \pm 57 \text{ Bq m}^{-2}$, decay-corrected to 2016).

5. Perspectives for future research

Beside the fact that baseline maps of ^{137}Cs fallout are of fundamental importance in case of future accidental radionuclide emissions, their use in geomorphological studies provides an opportunity to reconstruct soil redistribution due to soil erosion processes at larger scales (Chappell et al., 2011b; Meusburger et al., 2020). Although estimates of erosion rates at continental scales are debated (Fiener

and Auerswald, 2016; Panagos et al., 2015; Panagos et al., 2016), prediction of ^{137}Cs initial fallout at
kilometric scales offers the potential to increase our knowledge of soil erosion processes in catchments
through the use of the ^{137}Cs inventory technique and its upscaling (Lizaga et al., 2018). In general, the
spatial distribution of anthropogenic radionuclides in soils and their use as tracers of environmental
processes is of significant importance for Earth and atmospheric sciences (Bhandari, 1970; Everett et
al., 2008; Hirose, 2012; Igarashi et al., 2011; Jagercikova et al., 2017).

Considering that no radioactive fallout occurred since 1980 in South America, and as a consequence of
radioactive decay, ^{137}Cs activities in soils of South America will continue decreasing and become
increasingly difficult to measure without ultra-low background gamma spectrometry facilities (Evrard
et al., 2020). The development of surrogate tracers appears necessary for further geomorphological
studies using fallout radionuclides, especially in the Southern hemisphere that received only 23% of
the total bomb-fallout emitted worldwide according to UNSCEAR (2000). In the last several decades,
the efficiency of the $^{239+240}\text{Pu}$ inventory technique to quantify soil erosion rates has been demonstrated
in countries located in the Northern hemisphere (Alewell et al., 2014; Alewell et al., 2017; Meusbürger
et al., 2016). To the best of our knowledge, one study investigating soil erosion rates using ^{239}Pu was
carried out in Australia (Lal et al., 2020). In contrast to ^{137}Cs and $^{239+240}\text{Pu}$, the continuous input of fallout
radionuclides including excess lead-210 ($^{210}\text{Pb}_{\text{xs}}$) should be considered as an efficient tracer of soil
erosion, especially in regions where low ^{137}Cs fallout occurred (Evrard et al., 2020; Gaspar et al., 2013;
Porto et al., 2009; Walling et al., 2011). The effectiveness of these techniques in South America remains
unknown and it should be investigated to assess soil redistribution under climate and land use changes.

Overall, a significant lack of information about ^{137}Cs inventories in reference soil sites is observed in
areas of South America exposed to extensive clearcutting, overgrazing and cropping intensification. As
an example, central Brazil in general and the Cerrado Biome in particular (Fig. 9) should have received

sufficient levels of ^{137}Cs fallout to investigate soil redistribution rates induced by deforestation and land uses changes. A better characterization of ^{137}Cs fallout at the scale of South America is also required with the addition of soil inventory measurements in North-Western parts of South America, where some areas should have received sufficient fallout to conduct geomorphological studies despite what was previously expected from the UNSCEAR reports. Accordingly, future research should strive to ensure that basic information (e.g. rainfall databases used) and details on the sampling design (e.g. number of soil profiles used to estimate ^{137}Cs inventories in reference soil sites) are properly documented to improve future model predictions and better consider the issues of uncertainty and data reliability.

6. Conclusions

Based on a compilation of published information, additional measurements and rainfall data over the period 1950-2000, this work represents one of the first approaches to spatialize the reference levels of fallout ^{137}Cs at the subcontinental-scale of South America. The current research demonstrates that the ^{137}Cs inventories technique should be appropriate to assess soil redistribution rates during the agricultural intensification period in Chile, Argentina, Uruguay and Southern Brazil where detectable levels of ^{137}Cs can be expected to be found in sites exposed to erosion. This technique should theoretically be applicable in other countries where no information was available to date, such as Paraguay, Bolivia and Peru. Further investigations should be conducted in equatorial regions where information on ^{137}Cs fallout is scarce. Our results indicate that ^{137}Cs inventories in this region may be higher than expected from the UNSCEAR reports. Additional sampling is necessary to verify whether it will be either complicated or impossible to quantify soil erosion using bomb-derived ^{137}Cs in equatorial regions exposed to extensive clearcutting and agricultural expansion.

In addition to the priority complementary sampling locations identified through the use of conditioned

Latin Hypercube sampling (cLHS), the South American continent could be subdivided into regions corresponding to different biomes in which the fate of the global ^{137}Cs fallout is expected to be homogeneous. Consequently, future sampling campaigns should strive to ensure that all biomes are well covered with a sufficient number of soil profiles.

The map generated can be used both to validate ^{137}Cs inventories collected in the field or as a decision-support tool to guide the implementation of the ^{137}Cs technique in intensive agricultural landscapes of South America. This baseline map will also be particularly useful for a wide range of Earth science applications, including the vertical transfers in soils, the circulation of air masses and ocean currents. Of note, this map is provisional as it is only based on the data published until early 2020 and it can be optimized through the incorporation of additional ^{137}Cs inventory measurements in South America.

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7. References

- Alewell, C., Meusburger, K., Juretzko, G., Mabit, L. and Ketterer, M.E., 2014. Suitability of $^{239+240}\text{Pu}$ and ^{137}Cs as tracers for soil erosion assessment in mountain grasslands. *Chemosphere*, 103: 274-280.
- Alewell, C., Pitois, A., Meusburger, K., Ketterer, M. and Mabit, L., 2017. $^{239+240}\text{Pu}$ from “contaminant” to soil erosion tracer: Where do we stand? *Earth-Science Reviews*, 172: 107-123.
- Almgren, S., Nilsson, E., Erlandsson, B. and Isaksson, M., 2006. GIS supported calculations of ^{137}Cs deposition in Sweden based on precipitation data. *Science of the Total Environment*, 368(2-3): 804-813.

- Alonso, J., Audicio, P., Martinez, L., Scavone, M. and Rezzano, E., 2012. Comparison of measured ¹³⁷Cs data and USLE/RUSLE simulated long-term erosion rates. *Agrociencia-Sitio en Reparación*, 16(3): 261-267.
- Aoyama, M., Hirose, K. and Igarashi, Y., 2006. Re-construction and updating our understanding on the global weapons tests ¹³⁷Cs fallout. *Journal of Environmental Monitoring*, 8(4): 431.
- Ares, M.G., Bongiorno, F., Holzman, M., Chagas, C., Varni, M. and Entraigas, I., 2016. Water erosion and connectivity analysis during a year with high precipitations in a watershed of Argentina. *Hydrology Research*, 47(6): 1239-1252.
- Ayub, J.J., Velasco, R., Rizzotto, M., Quintana, E. and Aguiar, J., 2008. 40 K, 115 Cs and 226 Ra Soil and Plant Content in Seminatural Grasslands of Central Argentina, AIP Conference Proceedings. AIP, pp. 273-276.
- Bacchi, O.O.S., Reichard, K., Sparovek, G. and Ranieri, S.B.L., 2000. Soil erosion evaluation in a small watershed in Brazil through ¹³⁷Cs fallout redistribution analysis and conventional models. *Acta Geologica Hispanica*, 35(3-4): 251-259.
- Bakker, M.M., Govers, G., Jones, R.A. and Rounsevell, M.D.A., 2007. The Effect of Soil Erosion on Europe's Crop Yields. *Ecosystems*, 10(7): 1209-1219.
- Bakker, M.M., Govers, G. and Rounsevell, M.D., 2004. The crop productivity-erosion relationship: an analysis based on experimental work. *Catena*, 57(1): 55-76.
- Banfield, C.C., Braun, A.C., Barra, R., Castillo, A. and Vogt, J., 2018. Erosion proxies in an exotic tree plantation question the appropriate land use in Central Chile. *CATENA*, 161: 77-84.
- Becker, A.G., Moraes, B.S., Menezes, C.C., Loro, V.L., Santos, D.R., Reichert, J.M. and Baldisserotto, B., 2009. Pesticide contamination of water alters the metabolism of juvenile silver catfish, *Rhamdia quelen*. *Ecotoxicol Environ Saf*, 72(6): 1734-9.
- Bennett, B.G., 2002. Worldwide dispersion and deposition of radionuclides produced in atmospheric tests : Fallout from atmospheric nuclear tests : Impact on science and society. *Health Physics*, 82(5): 644-655.
- Bhandari, N., 1970. Transport of air in the stratosphere as revealed by radioactive tracers. *Journal of Geophysical Research*, 75(15): 2927-2930.
- Bouisset, P., Nohl, M., Bouville, A. and Leclerc, G., 2018. Inventory and vertical distribution of ¹³⁷Cs, ²³⁹⁺²⁴⁰Pu and ²³⁸Pu in soil from Raivavae and Hiva Oa, two French Polynesian islands in the southern hemisphere. *Journal of Environmental Radioactivity*, 183: 82-93.
- Buesseler, K.O. and Benitez, C.R., 1994. Determination of mass accumulation rates and sediment radionuclide inventories in the deep Black Sea. *Deep-Sea Research Part I-Oceanographic Research Papers*, 41(11-12): 1605-1615.
- Bujan, A., Santanatoglia, O.J., Chagas, C., Massobrio, M., Castiglioni, M., Yanez, M., Ciallella, H. and Fernandez, J., 2003. Soil erosion evaluation in a small basin through the use of Cs-137 technique. *Soil & Tillage Research*, 69(1-2): 127-137.
- Camargo, F.A.O., Silva, L.S., Merten, G.H., Carlos, F.S., Baveye, P.C. and Triplett, E.W., 2017. Brazilian Agriculture in Perspective. *Advances in Agronomy*, pp. 53-114.
- Cambray, R., Playford, K., Lewis, G. and Carpenter, R., 1989. Radioactive fallout in air and rain: results to the end of 1988. AERE R13575. Atomic Energy Authority, London, UK.
- Chappell, A., Hancock, G., Viscarra Rossel, R.A. and Loughran, R., 2011a. Spatial uncertainty of the ¹³⁷Cs reference inventory for Australian soil. *Journal of Geophysical Research*, 116(F4).
- Chappell, A., Viscarra Rossel, R.A. and Loughran, R., 2011b. Spatial uncertainty of ¹³⁷Cs-derived net (1950s–1990) soil redistribution for Australia. *Journal of Geophysical Research: Earth Surface*, 116(F4).
- Chartin, C., Evrard, O., Salvador-Blanes, S., Hinschberger, F., Van Oost, K., Lefèvre, I., Daroussin, J. and Macaire, J.-J., 2013. Quantifying and modelling the impact of land consolidation and field borders on soil redistribution in agricultural landscapes (1954–2009). *Catena*, 110: 184-195.
- Correchel, V., Bacchi, O.O.S., Reichardt, K. and De Maria, I.C., 2005. Random and systematic spatial variability of ¹³⁷Cs inventories at reference sites in South-Central Brazil. *Scientia Agricola*, 62(2): 173-178.

- de Tombeur, F., Cornu, S., Bourlès, D., Duvivier, A., Pupier, J., Aster, T., Brossard, M. and Evrard, O., 2020. Retention of ^{10}Be , ^{137}Cs and ^{210}Pb in soils: Impact of physico-chemical characteristics. *Geoderma*, 367.
- Didone, E.J., Minella, J.P.G., Schneider, F.J.A., Londero, A.L., Lefevre, I. and Evrard, O., 2019. Quantifying the impact of no-tillage on soil redistribution in a cultivated catchment of Southern Brazil (1964-2016) with Cs-137 inventory measurements. *Agriculture Ecosystems & Environment*, 284.
- Efron, B. and Tibshirani, R.J., 1994. An introduction to the bootstrap. CRC press.
- Ehnhalt, D., 1973. Turnover times of ^{137}Cs and HTO in the troposphere and removal rates of natural aerosol particles and water vapor. *Journal of Geophysical Research*, 78(30): 7076-7086.
- Everett, S., Tims, S., Hancock, G., Bartley, R. and Fifield, L.K., 2008. Comparison of Pu and ^{137}Cs as tracers of soil and sediment transport in a terrestrial environment. *Journal of Environmental Radioactivity*, 99(2): 383-393.
- Evrard, O., Bièdiers, C.L., Vandaele, K. and van Wesemael, B., 2007. Spatial and temporal variation of muddy floods in central Belgium, off-site impacts and potential control measures. *Catena*, 70(3): 443-454.
- Evrard, O., Chaboche, P.-A., Ramon, R., Foucher, A. and Laceby, J.P., 2020. A global review of sediment source fingerprinting research incorporating fallout radiocesium (^{137}Cs). *Geomorphology*: 107103.
- FAO, 2015. Status of the world's soil resources: main report. FAO : ITPS, Rome.
- FAO/IAEA, 2017. Use of ^{137}Cs for soil erosion assessment. Fulajtar, E., Mabit, L., Renschler, C.S., Lee Zhi Yi, A., . In: F.a.A.O.o.t.U. Nations (Editor), Rome, Italy, pp. 64.
- Fiener, P. and Auerwald, K., 2016. Comment on "The new assessment of soil loss by water erosion in Europe" by Panagos et al. (*Environmental Science & Policy* 54 (2015) 438–447). *Environmental Science & Policy*, 57: 140-142.
- Foucher, A., Salvador-Blanes, S., Evrard, O., Simonneau, A., Chapron, E., Courp, T., Cerdan, O., Lefèvre, I., Adriaensen, H., Lecompte, F. and Desmet, M., 2014. Increase in soil erosion after agricultural intensification: Evidence from a lowland basin in France. *Anthropocene*, 7: 30-41.
- Fukuyama, T., Takenaka, C. and Onda, Y., 2005. ^{137}Cs loss via soil erosion from a mountainous headwater catchment in central Japan. *Science of the total environment*, 350(1-3): 238-247.
- Furuichi, T. and Wasson, R.J., 2013. Caesium-137 in Southeast Asia: Is there enough left for soil erosion and sediment redistribution studies? *Journal of Asian Earth Sciences*, 77: 108-116.
- Garcia Agudo, E., 1998. Global distribution of Cs-137 inputs for soil erosion and sedimentation studies, In: Use of Caesium-137 in the Study of Soil Erosion and Sedimentation. IAEA TECDOC 1028., pp. 117-121.
- Garrett, R.D., Koh, I., Lambin, E.F., De Waroux, Y.I.P., Kastens, J.H. and Brown, J., 2018. Intensification in agriculture-forest frontiers: Land use responses to development and conservation policies in Brazil. *Global Environmental Change*, 53: 233-243.
- Gaspar, L., Navas, A., Walling, D., Machín, J. and Arozamena, J.G., 2013. Using ^{137}Cs and ^{210}Pb to assess soil redistribution on slopes at different temporal scales. *Catena*, 102: 46-54.
- Gerlach, R.W., Kowalski, B.R. and Wold, H.O., 1979. Partial least squares path modelling with latent variables, WASHINGTON UNIV SEATTLE LAB FOR CHEMOMETRICS.
- Gilbert, E.S., Land, C.E. and Simon, S.L., 2002. Health effects from fallout. *Health Physics*, 82(5): 726-735.
- Handl, J., Sachse, R., Jakob, D., Michel, R., Evangelista, H., Gonçalves, A.C. and de Freitas, A.C., 2008. Accumulation of ^{137}Cs in Brazilian soils and its transfer to plants under different climatic conditions. *Journal of Environmental Radioactivity*, 99(2): 271-287.
- Health and Safety Laboratory, E., New York 10014, 1977. Final tabulation of monthly strontium-90 fallout data : 1954-1976.
- Henry, A., Mabit, L., Jaramillo, R.E., Cartagena, Y. and Lynch, J.P., 2013. Land use effects on erosion and carbon storage of the Rio Chimbo watershed, Ecuador. *Plant and Soil*, 367(1-2): 477-491.

- Hijmans, R.J., Cameron, S.E., Parra, J.L., Jones, P.G. and Jarvis, A., 2005. Very high resolution interpolated climate surfaces for global land areas. *International Journal of Climatology*, 25(15): 1965-1978.
- Hijmans, R.J., Van Etten, J., Cheng, J., Mattiuzzi, M., Sumner, M., Greenberg, J.A., Lamigueiro, O.P., Bevan, A., Racine, E.B. and Shortridge, A., 2015. Package 'raster'. R package, 734.
- Hirose, K., 2012. Uranium, thorium and anthropogenic radionuclides as atmospheric tracers, *Handbook of Environmental Isotope Geochemistry*. Springer, pp. 591-611.
- Höskuldsson, A., 1988. PLS regression methods. *Journal of chemometrics*, 2(3): 211-228.
- Igarashi, Y., Fujiwara, H. and Jugder, D., 2011. Change of the Asian dust source region deduced from the composition of anthropogenic radionuclides in surface soil in Mongolia. *Atmospheric Chemistry and Physics*, 11(14): 7069.
- Jagercikova, M., Cornu, S., Bourlès, D., Evrard, O., Hatté, C. and Balesdent, J., 2017. Quantification of vertical solid matter transfers in soils during pedogenesis by a multi-tracer approach. *Journal of Soils and Sediments*, 17(2): 408-422.
- Keesstra, S.D., Bouma, J., Wallinga, J., Tittonell, P., Smith, P., Cerdà, A., Montanarella, L., Quinton, J.N., Pachepsky, Y., van der Putten, W.H., Bardgett, R.D., Moolenaar, S., Mol, G., Jansen, B. and Fresco, L.O., 2016. The significance of soils and soil science towards realization of the United Nations Sustainable Development Goals. *Soil*, 2(2): 111-128.
- Kuhn, M., 2008. Building predictive models in R using the caret package. *Journal of statistical software*, 28(5): 1-26.
- La Manna, L., Gaspar, L., Tarabini, M., Quijano, L. and Navas, A., 2019. Cs-137 inventories along a climatic gradient in volcanic soils of Patagonia: Potential use for assessing medium term erosion processes. *Catena*, 181.
- LaBrecque, J., Rosales, P. and Cordoves, P., 2001. Anomalously high values of cesium-137 in soils on the Peninsula de Paraguana (Venezuela). *Journal of Radioanalytical and Nuclear Chemistry*, 247(3): 563-566.
- Lal, R., 2015. Restoring Soil Quality to Mitigate Soil Degradation. *Sustainability*, 7(5): 5875-5895.
- Lal, R., Fifield, L., Tims, S., Wasson, R. and Howe, D., 2020. A study of soil erosion rates using ²³⁹Pu, in the wet-dry tropics of northern Australia. *Journal of environmental radioactivity*, 211: 106085.
- Le Roux, G., Duffa, C., Vray, F. and Renaud, P., 2010. Deposition of artificial radionuclides from atmospheric Nuclear Weapon Tests estimated by soil inventories in French areas low-impacted by Chernobyl. *J Environ Radioact*, 101(3): 211-8.
- Liddicoat, C., Maschmedt, D., Clifford, D., Searle, R., Herrmann, T., Macdonald, L.M. and Baldock, J., 2015. Predictive mapping of soil organic carbon stocks in South Australia's agricultural zone. *Soil Research*, 53(8): 956-973.
- Lizaga, I., Quijano, L., Gaspar, L. and Navas, A., 2018. Estimating soil redistribution patterns with ¹³⁷Cs measurements in a Mediterranean mountain catchment affected by land abandonment. *Land Degradation & Development*, 29(1): 105-117.
- Loughran, R., Pennock, D. and Walling, D., 2002. Spatial distribution of caesium-137, *Handbook for the assessment of soil erosion and sedimentation using environmental radionuclides*. Springer, pp. 97-109.
- Mabit, L., Bernard, C., Lee Zhi Yi, A., Fulajtar, E., Dercon, G., Zaman, M., Toloza, A. and Heng, L., 2018. Promoting the use of isotopic techniques to combat soil erosion: An overview of the key role played by the SWMCN Subprogramme of the Joint FAO/IAEA Division over the last 20 years. *Land Degradation & Development*, 29(9): 3077-3091.
- Mabit, L., Bernard, C., Makhoulouf, M. and Laverdière, M., 2008. Spatial variability of erosion and soil organic matter content estimated from ¹³⁷Cs measurements and geostatistics. *Geoderma*, 145(3-4): 245-251.
- Mabit, L., Meusburger, K., Fulajtar, E. and Alewell, C., 2013. The usefulness of ¹³⁷Cs as a tracer for soil erosion assessment: A critical reply to Parsons and Foster (2011). *Earth-Science Reviews*, 127: 300-307.

- Macêdo, Í.L., 2009. Estudo de modelos em ambiente de geoprocessamento para a previsão de erosão e assoreamento de reservatórios: O caso da bacia do Rio Indaia – Uhe Três Marias, Mg. 340.
- Malakhov, S.G. and Pudovkina, I.B., 1970. Strontium 90 fallout distribution at middle latitudes of the Northern and Southern Hemispheres and its relation to precipitation. *Journal of Geophysical Research*, 75(18): 3623-3628.
- Malone, B., 2015. ithir: Soil data and some useful associated functions. R package version, 1.
- Martin, P. and McBride, J.L., 2012. Radionuclide Behaviour and Transport in the Tropical Atmospheric Environment, *Radioactivity in the Environment*. Elsevier, pp. 59-91.
- McBratney, A.B., Santos, M.M. and Minasny, B., 2003. On digital soil mapping. *Geoderma*, 117(1-2): 3-52.
- McCarthy, M., 1997. Nuclear bomb test fallout may cause many US cancers. *The Lancet*, 350(9075): 415.
- Mendez, M.J. and Buschiazzi, D.E., 2010. Wind erosion risk in agricultural soils under different tillage systems in the semiarid Pampas of Argentina. *Soil & Tillage Research*, 106(2): 311-316.
- Meusburger, K., Evrard, O., Alewell, C., Borrelli, P., Cinelli, G., Ketterer, M., Mabit, L., Panagos, P., van Oost, K. and Ballabio, C., 2020. Plutonium aided reconstruction of caesium atmospheric fallout in European topsoils. *Scientific Reports*, 10(1).
- Meusburger, K., Mabit, L., Ketterer, M., Park, J.-H., Sandor, T., Porto, P. and Alewell, C., 2016. A multi-radionuclide approach to evaluate the suitability of ²³⁹⁺²⁴⁰Pu as soil erosion tracer. *Science of the Total Environment*, 566: 1489-1499.
- Minasny, B. and McBratney, A.B., 2006. A conditioned Latin hypercube method for sampling in the presence of ancillary information. *Computers & Geosciences*, 32(9): 1378-1388.
- Minella, J.P., Walling, D.E. and Merten, G.H., 2014. Establishing a sediment budget for a small agricultural catchment in southern Brazil, to support the development of effective sediment management strategies. *Journal of Hydrology*, 519: 2189-2201.
- Montes, M., Silva, L., Sá, C., Runco, J., Taylor, M. and Desimoni, J., 2013. Inventories and concentration profiles of ¹³⁷Cs in undisturbed soils in the northeast of Buenos Aires Province, Argentina. *Journal of environmental radioactivity*, 116: 133-140.
- Montgomery, D.R., 2007. Soil erosion and agricultural sustainability. *Proceedings of the National Academy of Sciences*, 104(33): 13268-13272.
- Navas, A., Machín, J. and Soto, J., 2005. Assessing soil erosion in a Pyrenean mountain catchment using GIS and fallout ¹³⁷Cs. *Agriculture, Ecosystems & Environment*, 105(3): 493-506.
- Navas, A., Serrano, E., López-Martínez, J., Gaspar, L. and Lizaga, I., 2018. Interpreting environmental changes from radionuclides and soil characteristics in different landform contexts of Elephant Island (maritime Antarctica). *Land Degradation & Development*, 29(9): 3141-3158.
- Owens, P., Batalla, R., Collins, A., Gomez, B., Hicks, D., Horowitz, A., Kondolf, G., Marden, M., Page, M. and Peacock, D., 2005. Fine-grained sediment in river systems: environmental significance and management issues. *River research and applications*, 21(7): 693-717.
- Owens, P.N. and Walling, D.E., 1996. Spatial variability of caesium-137 inventories at reference sites: an example from two contrasting sites in England and Zimbabwe. *Applied Radiation and Isotopes*, 47(7): 699-707.
- Palsson, S.E., Howard, B.J. and Wright, S.M., 2006. Prediction of spatial variation in global fallout of ¹³⁷Cs using precipitation. *Sci Total Environ*, 367(2-3): 745-56.
- Panagos, P., Borrelli, P., Poesen, J., Ballabio, C., Lugato, E., Meusburger, K., Montanarella, L. and Alewell, C., 2015. The new assessment of soil loss by water erosion in Europe. *Environmental Science & Policy*, 54: 438-447.
- Panagos, P., Borrelli, P., Poesen, J., Meusburger, K., Ballabio, C., Lugato, E., Montanarella, L. and Alewell, C., 2016. Reply to the comment on “The new assessment of soil loss by water erosion in Europe” by Fiener & Auerswald. *Environmental Science & Policy*, 57: 143-150.
- Parsons, A.J. and Foster, I.D.L., 2011. What can we learn about soil erosion from the use of ¹³⁷Cs? *Earth-Science Reviews*, 108(1-2): 101-113.

- Parsons, A.J. and Foster, I.D.L., 2013. The assumptions of science. *Earth-Science Reviews*, 127: 308-310.
- Pebesma, E.J., 2018. Simple features for R: Standardized support for spatial vector data. *R J.*, 10(1): 439.
- Pennock, D., 2000. Suitability of ¹³⁷Cs redistribution as an indicator of soil quality. *Acta geológica hispánica*: 213-217.
- Porto, P., Walling, D.E., Callegari, G. and Capra, A., 2009. Using caesium-137 and unsupported lead-210 measurements to explore the relationship between sediment mobilisation, sediment delivery and sediment yield for a Calabrian catchment. *Marine and Freshwater research*, 60(7): 680-689.
- Quinton, J.N., Govers, G., Van Oost, K. and Bardgett, R.D., 2010. The impact of agricultural soil erosion on biogeochemical cycling. *Nature Geoscience*, 3(5): 311-314.
- Racoviteanu, A., 2007. GLIMS Glacier Database, Version 1. Analysis IDs 60827-61235, Accessed 1/17/2017.
- Ritchie, J. and Ritchie, C., 2007. Bibliography of Publications of ¹³⁷Cesium Studies Related to Erosion and Sediment Deposition. Hydrology and Remote Sensing Laboratory, Beltsville, USA.
- Ritchie, J.C. and McHenry, J.R., 1990. Application of Radioactive Fallout Cesium-137 for Measuring Soil Erosion and Sediment Accumulation Rates and Patterns: A Review. *Journal of Environment Quality*, 19(2): 215.
- Rossel, R.V., Chen, C., Grundy, M., Searle, R., Clifford, D. and Campbell, P., 2015. The Australian three-dimensional soil grid: Australia's contribution to the GlobalSoilMap project. *Soil Research*, 53(8): 845-864.
- Roudier, P., 2011. *clhs: a R package for conditioned Latin hypercube sampling*. R package version 0.5-1.
- Schuller, P., Bunzl, K., Voigt, G., Ellies, A. and Castillo, A., 2003a. Global fallout ¹³⁷Cs accumulation and vertical migration in selected soils from South Patagonia. *Journal of Environmental Radioactivity*, 71(1): 43-60.
- Schuller, P., Ellies, A., Castillo, A. and Salazar, I., 2003b. Use of ¹³⁷Cs to estimate tillage- and water-induced soil redistribution rates on agricultural land under different use and management in central-south Chile. *Soil & Tillage Research*, 69(1-2): 69-83.
- Schuller, P., Voigt, G., Handl, J., Ellies, A. and Oliva, L., 2002. Global weapons' fallout ¹³⁷Cs in soils and transfer to vegetation in south-central Chile. *Journal of Environmental Radioactivity*, 62(2): 181-193.
- Simon, S.L., Bouville, A. and Land, C.E., 2006. Fallout from nuclear weapons tests and cancer risks: exposures 50 years ago still have health implications today that will continue into the future. *American Scientist*, 94(1): 48-57.
- Steffen, W., Broadgate, W., Deutsch, L., Gaffney, O. and Ludwig, C., 2015. The trajectory of the Anthropocene: the great acceleration. *The Anthropocene Review*, 2(1): 81-98.
- Steinhauser, G., Brandl, A. and Johnson, T.E., 2014. Comparison of the Chernobyl and Fukushima nuclear accidents: a review of the environmental impacts. *Sci Total Environ*, 470-471: 800-17.
- Tagami, K., Tsukada, H. and Uchida, S., 2019. Quantifying spatial distribution of ¹³⁷Cs in reference site soil in Asia. *CATENA*, 180: 341-345.
- Tamura, T., 1961. Cesium sorption reactions as indicator of clay mineral structures. *Clays and Clay Minerals*, 10(1): 389-398.
- Tamura, T. and Jacobs, D., 1960. Structural implications in cesium sorption. *Health physics*, 2(4): 391-398.
- Tassano, M., Montañez, A., Nuñez, L., Trasante, T., González, J., Irigoyen, J., Cabral, P. and Cabrera, M., 2020. Spatial cross-correlation between physicochemical and microbiological variables at superficial soil with different levels of degradation. *CATENA*: 105000.
- Team R Core, 2013. *R: A language and environment for statistical computing*. Vienna, Austria.
- Tenenhaus, M., 1998. *La régression PLS: théorie et pratique*. Editions technip.

- Turney, C.S.M., Palmer, J., Maslin, M.A., Hogg, A., Fogwill, C.J., Southon, J., Fenwick, P., Helle, G., Wilmschurst, J.M., McGlone, M., Bronk Ramsey, C., Thomas, Z., Lipson, M., Beaven, B., Jones, R.T., Andrews, O. and Hua, Q., 2018. Global Peak in Atmospheric Radiocarbon Provides a Potential Definition for the Onset of the Anthropocene Epoch in 1965. *Scientific Reports*, 8(1): 3293.
- UNSCEAR, 1962. Report of the United Nations Scientific Committee on the effects of atomic radiation. Annex F : Environmental contamination, New York.
- UNSCEAR, 2000. Effects of Ionizing Radiation. United Nations, New York: 453-487.
- UNSCEAR, 2008. Sources and effects of ionizing radiation: United Nations Scientific Committee on the Effects of Atomic Radiation: UNSCEAR 2008 report to the General Assembly, with scientific annexes. United Nations, New York.
- Vanwalleghem, T., Gómez, J.A., Infante Amate, J., González de Molina, M., Vanderlinden, K., Guzmán, G., Laguna, A. and Giráldez, J.V., 2017. Impact of historical land use and soil management change on soil erosion and agricultural sustainability during the Anthropocene. *Anthropocene*, 17: 13-29.
- Volchok, H., 1965. Distribution of strontium-90 in surface air during 1963. *Nature*, 206(4988): 1031-1033.
- Volchok, H., 1966. The global strontium 90 budget. *Journal of Geophysical Research*, 71(6): 1515-1518.
- Wadoux, A.M.-C., Brus, D.J. and Heuvelink, G.B., 2019. Sampling design optimization for soil mapping with random forest. *Geoderma*, 355: 113913.
- Walling, D. and He, Q., 2000. The global distribution of bomb-derived ¹³⁷Cs reference inventories. Final report on IAEA technical contract, 10361: 1-11.
- Walling, D., Zhang, Y. and He, Q., 2011. Models for deriving estimates of erosion and deposition rates from fallout radionuclide (caesium-137, excess lead-210, and beryllium-7) measurements and the development of user friendly software for model implementation. Impact of soil conservation measures on erosion control and soil quality. IAEA-TECDOC-1665: 11-33.
- Walling, D.E. and Quine, T., 1990. Calibration of caesium-137 measurements to provide quantitative erosion rate data. *Land Degradation & Development*, 2(3): 161-175.
- Wei, T., Simko, V., Levy, M., Xie, Y., Jin, Y. and Zemla, J., 2017. Package 'corrplot'. *Statistician*, 56(316): e24.
- Wickham, H., 2016. *ggplot2: elegant graphics for data analysis*. springer.
- Wingeyer, A.B., Amado, T.J.C., Perez-Bidegain, M., Studdert, G.A., Varela, C.H.P., Garcia, F.O. and Karlen, D.L., 2015. Soil Quality Impacts of Current South American Agricultural Practices. *Sustainability*, 7(2): 2213-2242.
- Wright, S.M., Howard, B.J., Strand, P., Nylén, T. and Sickel, M.A.K., 1999. Prediction of ¹³⁷Cs deposition from atmospheric nuclear weapons tests within the Arctic. *Environmental Pollution*, 104(1): 131-143.
- Zapata, F., 2002. Handbook for the assessment of soil erosion and sedimentation using environmental radionuclides, 219. Springer.
- Zapata, F., 2003. The use of environmental radionuclides as tracers in soil erosion and sedimentation investigations: recent advances and future developments. *Soil and Tillage Research*, 69(1-2): 3-13.

Figure 1: Spatial distribution of ^{137}Cs reference soil sites in undisturbed soil profiles of South America as documented in the literature and collected by the authors.

Figure 2: Variation of ^{137}Cs inventories in reference soil sites (Bq m⁻²), decay-corrected to 2020 and plotted against south latitude.

Figure 3: Distribution of mean ^{137}Cs in reference soil sites documented in our review for each 10° latitude bands compared with the distribution of radionuclide fallout from thermonuclear bomb testing with latitude, after UNSCEAR (2000). This distribution was calculated with 5000 samples generated using bootstrap iterations; the circle is the calculated mean while the whiskers represent the 95% confident interval.

Figure 4: Relationships between mean annual precipitation and ¹³⁷Cs inventories in reference soil sites for different latitude ranges.

Figure 5: Results of the multiple regressions using latitude, longitude, elevation and rainfall plotted with the 95% confidence intervals.

Figure 6: Correlogram plot for ^{137}Cs inventories in reference soil sites, mean monthly/annual precipitation rates (1950-2000) and geographical information. Significant levels > 0.1 are represented as white squares.

Figure 7: Baseline ^{137}Cs inventories in reference soil sites (Bq m^{-2} , 2020) estimated by Partial Least Square Regression (PLSR) with a spatial resolution of 2 km. Black dots represent those ^{137}Cs inventories compiled in the literature and measured by the authors, while green dots represent the additional samples that should be analysed in the future as determined by conditioned Latin Hypercube Sampling (cLHS) to improve model quality. Areas above 1,800 m a.s.l., glaciers and large water bodies have been masked in white using the GMTED2010 30-arc-second elevation database (Danielson and Gesch, 2011) and the GLIMS glacier database (Racoviteanu, 2007).

Figure 8: Prediction uncertainties of the baseline map of ^{137}Cs inventories in reference soil sites assessed through a non-parametric bootstrap approach ($n=100$ contributing predictions). Areas above 1,800 m a.s.l., glaciers and large water bodies have been masked in white using the GMTED2010 30-arc-second elevation database (Danielson and Gesch, 2011) and the GLIMS glacier database (Racoviteanu, 2007).

Figure 9: (A) Potential use of ^{137}Cs fallout inventories for Earth Science applications in South America. Areas above 1,800 m a.s.l., glaciers and large water bodies have been masked in white using the GMTED2010 30-arc-second elevation database (Danielson and Gesch, 2011) and the GLIMS glacier database (Racoviteanu, 2007). (B) cropland distribution across South America in a nominal 30-meter resolution (GFSAD30 Project), and (C) terrestrial biomes of South America (Dinerstein et al., 2017)