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1 **Mapping the spatial distribution of global ¹³⁷Cs fallout in soils of South America as a baseline for**
2 **Earth Science studies**

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25 **Abstract**

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3 26 Owing to the rapid expansion of agriculture in South America in recent decades, soil erosion and fine
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5 27 sediment supply to river networks, which lead to deleterious on-site and off-site environmental
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7 28 impacts, are exacerbated in intensively cultivated catchments. Measuring soil inventories of bomb-
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9 29 derived fallout radiocesium (^{137}Cs) bound to fine particles is one of the few techniques available to
10
11 30 reconstruct soil redistribution rates and evaluate the sustainability of farming practices over the recent
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13 31 phase of agricultural intensification (1960s–2020). However, information about the spatial distribution
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15 32 of ^{137}Cs fallout across the soils of South America remains scarce, and the published data has not been
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17 33 synthesized at the scale of this subcontinent so far. The objective of the current research is therefore
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19 34 to quantify and map the initial ^{137}Cs fallout at the scale of South America, based on the compilation of
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21 35 published ^{137}Cs inventories, additional measurements conducted on undisturbed soil profiles and
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23 36 digital soil mapping as this baseline information may be useful for a wide range of Earth Science
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25 37 applications. A database of ^{137}Cs inventories at 96 reference sites (i.e. areas without soil erosion nor
26
27 38 accumulation) has been compiled for a variety of soil profiles (Argentina = 10, Brazil = 34, Chile = 46,
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29 39 Uruguay = 5, French Guiana = 1) located between 5.3° North latitude and 53° South latitude. The spatial
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31 40 distribution of ^{137}Cs fallout was shown to be highly latitude-dependent, with a maximum in the 30-50°
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33 41 South latitude band. There were higher fallout levels than expected between 20 to 60° South latitude
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35 42 compared to the previous estimations made by UNSCEAR. A partial least square regression approach
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37 43 based on rainfall data and geographical information as covariates was used to create a baseline map
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39 44 of ^{137}Cs fallout in soils of continental South America. This baseline map provides a powerful reference
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41 45 dataset to anticipate the order of magnitude of ^{137}Cs inventories in undisturbed soil profiles collected
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43 46 in Brazil and Southern Chile and for numerous other applications in Earth Sciences. The potential
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45 47 application of the ^{137}Cs inventory technique in countries of South America in general, and in regions
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47 48 with vulnerable ecosystems threatened by the expansion of agricultural activities in particular, is
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49 49 discussed in light of this comprehensive literature review. Furthermore, the regions (i.e. the North-
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51 50 Western part of the continent) where additional samples should be collected in priority to improve
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53 51 this baseline map are outlined. Our results demonstrate that ^{137}Cs inventories are sufficiently high to
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1
2 52 investigate soil redistribution rates in most of South American countries where detectable levels of
3 53 ^{137}Cs can be expected to be found in sites exposed to erosion.
4

5 54 **Keywords:** Digital soil mapping, Caesium-137, Soil erosion, Soil redistribution rates, Reference soil sites
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10 11 56 **1. Introduction** 12 13 57

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15 58 Soil is essentially a non-renewable resource over the human timescale (Lal, 2015). The future use of
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17 59 this valuable resource is threatened by anthropogenic activities such as the intensification of
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19 60 agricultural practices (Keesstra et al., 2016). The Great Acceleration period that started during the
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21 61 second half of the 20th century induced a significant increase of soil erosion and degradation (Steffen
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23 62 et al., 2015), leading to extensive on-site and off-site impacts. On-site, soil erosion threatens soil
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25 63 fertility and agricultural yields (Bakker et al., 2007; Bakker et al., 2004; Vanwalleghem et al., 2017).
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27 64 Fine-grained sediment transported off-site, which may be associated with nutrients and contaminants,
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29 65 leads to the reduction of water quality, the degradation of aquatic habitats and reservoir siltation
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31 66 (Becker et al., 2009; Evrard et al., 2007; Foucher et al., 2014). Fine sediment supply induced by soil
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33 67 erosion processes is exacerbated in intensively cultivated catchments, leading to deleterious
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35 68 consequences for river systems (Owens et al., 2005) and to the disturbance of global biogeochemical
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37 69 cycles (Quinton et al., 2010).
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44 70 In order to evaluate the sustainability of agricultural practices and promote effective soil management
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46 71 practices, a fundamental prerequisite is to quantify soil erosion rates. Measuring soil inventories of
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48 72 fallout radionuclide Caesium-137 (^{137}Cs , $t_{1/2} = 30.2$ years) has shown great potential to provide
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50 73 retrospective information on soil redistribution rates over the medium-term (Ritchie and Ritchie,
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52 74 2007), despite the critiques made on this method (Parsons and Foster, 2011). The latter authors
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54 75 recommended that future studies should present their results in light of the underlying hypotheses
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56 76 (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).

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

102 10-degree latitudinal bands was established in the UNSCEAR report published in 2000. In parallel, soil
1 erosion and soil-to-crop transfer studies have provided more accurate local information on the spatial
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3 erosion and soil-to-crop transfer studies have provided more accurate local information on the spatial
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5 distribution of initial ^{137}Cs fallout worldwide (Owens and Walling, 1996; Ritchie and McHenry, 1990;
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8 Walling and He, 2000). The deposition of fallout radionuclides onto the Earth surface is conditioned by
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11 two major factors: latitude and rainfall. Ground deposition of radionuclides from the global fallout was
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13 shown to be highly latitude-dependent, with a maximum in the 40-50° bands in both hemispheres
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15 (UNSCEAR, 2000). In addition, the activity of ^{137}Cs deposited on the ground and its variation from one
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17 location to another is closely linked to the annual rainfall rates, as a consequence of precipitation
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20 scavenging (Bouisset et al., 2018; La Manna et al., 2019; Le Roux et al., 2010; Malakhov and Pudovkina,
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22 1970). Moreover, relationships between monthly rainfall rates and ^{90}Sr fallout were observed in South
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24 America, with a peak during the months of June-August (Volchok, 1965; Volchok, 1966). Based on these
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27 two factors, digital mapping using geostatistical approaches has been conducted at different scales to
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29 map bomb-derived ^{137}Cs fallout (Almgren et al., 2006; Aoyama et al., 2006; Chappell et al., 2011a;
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31 115
32 Furuichi and Wasson, 2013; Meusburger et al., 2020; Palsson et al., 2006). Although reconstructions
33 116
34 of baseline ^{137}Cs inventories were made for Europe, Australia and Eastern Asia, information about the
35 117
36 spatial distribution of ^{137}Cs in soils of South America remains scarce, which requires further
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39 investigation to determine the potential for application of ^{137}Cs for Earth Science studies in this
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41 subcontinent. Of note, the ^{137}Cs inventories in soils of the Southern hemisphere are usually sufficient
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43 to be measurable by gamma spectrometry when using appropriate low-background detectors during
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46 sufficient counting times (FAO/IAEA, 2017).

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49 Quantifying ^{137}Cs inventories in soils and calculating the corresponding soil redistribution rates during
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51 the intensive cultivation period (1960 to present) is particularly important to evaluate the sustainability
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54 of agricultural practices implemented in South America during the last several decades to maintain
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56 erosion at sustainable levels (Minella et al., 2014). During the last 50 years, most countries of the
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58 subcontinent have abandoned conventional tillage to implement no-till farming (Montgomery, 2007).
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61 This change in practice has been accompanied by an increase in the size of cultivated areas and their
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128 overall productivity (Wingeyer et al., 2015). However, no-till farming has been widely implemented as
129 a single conservation measure without additional practices to reduce soil loss, such as crop rotation or
130 contour cropping and terracing (Didone et al., 2019; Montgomery, 2007). As a consequence, soil
131 erosion by water remains the main soil degradation process in agricultural land of South America and
132 its real magnitude remains debated among agricultural and scientific communities (FAO, 2015).

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

145

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

2.1. Global fallout following nuclear weapon tests

152 The UNSCEAR reports provide the main source of information regarding ^{137}Cs deposition worldwide.
153 Early studies about the production and transport of nuclear weapon debris started in 1962 (UNSCEAR,
154 1962). It is currently accepted that a total of 502 atmospheric tests, with a total fission and fusion yield

155 of 440 Mt, were conducted from the mid-1950s to 1980 (UNSCEAR, 2008). Following the extensive
1 tests of atmospheric nuclear devices in 1962, the deposition of ¹³⁷Cs reached a peak in 1963 and 1965
2 156 tests of atmospheric nuclear devices in 1962, the deposition of ¹³⁷Cs reached a peak in 1963 and 1965
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4 157 in the Northern and Southern hemispheres, respectively (Cambray et al., 1989; Turney et al., 2018).
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7 158 The signing of the Partial Test-Ban Treaty on August 5, 1963 by the United Kingdom, the Soviet Union
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9 159 and the United States has contributed to the reduction of global emissions of artificial radionuclides
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11 160 into the environment. In soils of South America, no trace of Chernobyl and Fukushima Daichii-derived
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13 161 fallout radionuclides were detected, which makes the atmospheric weapon test fallout the single
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15 162 significant source of artificial radioactivity in this part of the world (Steinhauser et al., 2014).
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19 163 The radioactive debris emitted as a result of atmospheric nuclear tests were distributed between the
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21 164 surface of the ground or water and the tropospheric and stratospheric regions, depending on the type
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23 165 of test (offshore barge, top of a tower, under a tethered balloon, etc.), the location (altitude and firing
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25 166 latitude) and the power (kilotonnic or megatonnic). The majority of the radioactive debris was
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27 167 dispersed into the stratosphere (referred to as “stratospheric fallout” or “global” fallout). The activity
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29 168 that did not reach the stratosphere is referred to as “local / regional” fallout and “tropospheric fallout”
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31 169 (UNSCEAR, 2000).
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36 170 Local fallout includes radioactive aerosol particles generally larger than 50 µm in size that are
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38 171 deposited within a radius of about 100 km from the epicentre of the explosion (Garcia Agudo, 1998).
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40 172 Tropospheric fallout is characterized by smaller aerosols that were deposited with a mean atmospheric
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42 173 residence time of up to 30 days. During this period, the debris were dispersed within the latitude band
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44 174 of the initial injection and followed wind-driven trajectories, before falling to the ground as a
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46 175 consequence of precipitation scavenging (Bennett, 2002). Stratospheric fallout, which makes up a large
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48 176 portion of total deposition, consists of particles that are transported into the stratosphere, dispersed
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50 177 and then deposited globally, most of which occurs in the hemisphere of the initial injection.
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52 178 Stratospheric deposition accounts for the majority of global long-lived fission product residues
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54 179 (UNSCEAR, 2000).
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180 In order to estimate radionuclide deposition following nuclear detonations, two long-term monitoring
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2 181 programmes were established during the nuclear weapon test period. The first programme, conducted
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4 182 by the United Kingdom Atomic Energy Authority (UKAEA) consisted of 8 stations located in the United
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7 183 Kingdom and 18 stations installed in the rest of the world. The second programme, conducted by the
8
9 184 Environmental Measurements Laboratory in the United States (EML), as early as in 1954, consisted of
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11 185 177 stations distributed worldwide. This monitoring programme, based on monthly ^{90}Sr fallout
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13 186 sampling, was the largest and the most widely distributed and therefore, it has been adopted by the
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15
16 187 UNSCEAR to estimate the total hemispheric annual deposition of ^{90}Sr (UNSCEAR, 1993). The global
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18 188 distribution of ^{137}Cs was assessed assuming a $^{137}\text{Cs}/^{90}\text{Sr}$ fission yield ratio of 1.5, and a uniform
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21 189 distribution model for each 10-degree latitude band was published in the UNSCEAR (2000) report.
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24 190 The latitudinal distribution of ^{137}Cs described by UNSCEAR should be taken with caution as 50% of the
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26 191 data is missing from this monitoring programme (Evrard et al., 2020). Indeed, a significant proportion
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28 192 of data is lacking for monitoring stations ($n=34$) located in South America. In all, 59% of monthly ^{90}Sr
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30 193 fallout data is missing between 1954-1976, as most of the monitoring stations did not have continuous
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32 194 records (Health and Safety Laboratory, 1977). In addition, when studies investigating ^{137}Cs inventories
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34 195 in undisturbed soil profiles are conducted at regional or continental scales, the proportion of global
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36 196 fallout estimated by UNSCEAR appears to be underestimated (Aoyama et al., 2006; Chappell et al.,
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38 197 2011a; Schuller et al., 2002; Tagami et al., 2019). Regarding the information currently available for ^{137}Cs
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40 198 fallout at the scale of South America, the only reference is a study published by Garcia Agudo (1998)
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42 199 who also used results of these monitoring programmes to map the global inventory of ^{137}Cs from
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44 200 nuclear tests worldwide. Given the wide range of applications of ^{137}Cs in the scientific literature, as
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46 201 well as the debate raised on its use as a tracer for soil erosion assessment (Mabit et al., 2013; Parsons
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48 202 and Foster, 2011; Parsons and Foster, 2013), a more accurate characterization of the initial fallout of
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50 203 bomb-derived ^{137}Cs and its spatial distribution across South America is fundamental to provide a
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52 204 baseline for future studies.
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3 206 2.2. Data collection on ¹³⁷Cs in reference soil sites of South America

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9 208 A literature survey was carried out using Web of Science databases on March 1, 2020. Most data were

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11 209 found in peer-reviewed scientific articles published in English and Portuguese languages (n=17),

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13 210 although inventories were also taken from unpublished PhD manuscripts (n=2). The search keywords

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15 211 'soil erosion', 'cesium-137', '¹³⁷Cs' and 'inventorie(s)' were used in isolation and/or combination with

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17 212 additional keywords containing the country names of South America. In addition, six scientific articles

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19 213 that were not identified during the initial Web of Science search were included in the literature

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21 214 compilation during the review process. Three conditions were necessary to accept data: (1) the data

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23 215 were provided in Bq m⁻², (2) sampling locations were provided or easy to obtain and (3) information

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25 216 about sampling procedures was detailed. The sampling year or the year of decay-correction were also

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27 217 required to standardise all the data compiled. Seven studies did not mention the sampling year or the

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29 218 year of decay-correction. In this case, we made the assumption that sampling was conducted 4 years

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31 219 before the report publication, corresponding to the mean duration between sampling and publication

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33 220 in the current literature survey. All the data was decay-corrected to 2020 according to Eq. (1)

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

43

44 222 where λ is the decay constant of ¹³⁷Cs ($\lambda = \text{Ln}2/30.2 \text{ y}$) and t is the time (year) since the sampling year

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46 223 as reported in the corresponding article or estimated following the above-mentioned method. Each

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48 224 entry in the database corresponds to one undisturbed soil profile for which the ¹³⁷Cs inventory has

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50 225 been calculated, its geographical coordinates (latitude, longitude, elevation), annual rainfall rates,

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52 226 sampling method, ¹³⁷Cs inventory and the associated standard deviation estimated by the author and

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54 227 the associated decay-corrected values calculated from Eq. (1). In total, a database of 103 ¹³⁷Cs

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56 228 inventories at reference soil sites has been compiled, in four different countries and one overseas

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229 department of France (Antarctica = 2, Argentina = 10, Brazil = 38, Chile = 50, Uruguay = 2, French Guiana
1
2 230 = 1) between 5.3° North latitude and 62° South latitude and from 109.3° to 34.9° West longitude (Fig.
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4 231 1). Mean annual precipitation, the year of decay-correction, altitude and standard deviation were
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7 232 reported for 98, 82, 71 and 40 ¹³⁷Cs inventories calculated at these reference soil sites, respectively.
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9 233 Four ¹³⁷Cs inventories from Easter Island (Pacific Ocean) and Antarctica were removed as they were
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11 234 not obtained on the main South-American continent. Of note, four inventories obtained near the
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13 235 Amazon River (0 and 1 Bq m⁻² respectively) and in Southern Brazil (5 and 90 Bq m⁻²), analysed by Handl
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15 236 et al. (2008) may have been sampled in erosional areas rather than at reference sites, which may have
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17 237 an impact on the results of the current research. Conversely, the highest ¹³⁷Cs inventories were found
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19 238 in the central part of Chile (3113 and 2860 Bq m⁻²), at 40° South latitude (Schuller et al., 2002). These
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21 239 inventories, with elevated ¹³⁷Cs concentrations, may have been sampled in accumulation areas instead
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23 240 of reference locations. These six samples were considered as anomalous.
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29 241 To gain insight into the accuracy of the baseline map of ¹³⁷Cs fallout derived from this dataset,
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31 242 additional unpublished ¹³⁷Cs inventories in undisturbed soil profiles were used in the present study. In
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33 243 total, 24 soil cores were collected by co-authors in Uruguay down to a soil depth of 20cm to
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35 244 characterize three reference sites. All soil samples were collected using an Eijkelkamp soil sampler (5
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37 245 cm diameter) and then stored in polyethylene bags. Samples were ground and passed through a 2 mm
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39 246 sieve, then placed in 250 mL plastic Marinelli beakers. Gamma-ray measurements of ¹³⁷Cs (662 keV)
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41 247 were obtained by high purity germanium detector (HPGe) (Canberra) at Laboratorio de Radioquímica
42
43 248 (Centro de Investigaciones Nucleares, Universidad de la República, Montevideo, Uruguay) and at
44
45 249 Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). In summary,
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47 250 among the 103 ¹³⁷Cs inventories compiled in the literature and the additional three ¹³⁷Cs inventories
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49 251 collected by the co-authors, 96 ¹³⁷Cs inventories (Table 1) were used to perform the statistical analysis
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51 252 presented below.
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254 2.3. Spatial distribution of ¹³⁷Cs inventories in South America

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256 According to the current literature review, the highest ¹³⁷Cs inventories in reference soil sites were
257 found in the latitude band between 40 to 50° South (Fig. 2). From this location, inventories decreased
258 towards both the Equator and Southern Patagonia, where the mean ¹³⁷Cs inventories were 97 and 279
259 Bq m⁻², respectively. The distribution of ¹³⁷Cs inventories in reference soil sites of South America is
260 highly latitude-dependent, with a maximum in the 30-50° South latitude band, which is slightly higher
261 than that proposed by UNSCEAR for that same latitude range. In contrast, the ¹³⁷Cs inventories in the
262 0-20° South latitude band found in the literature is slightly lower than that proposed by UNSCEAR (Fig.
263 3). Attention should be paid as published inventories were not found in the literature for the North-
264 Western part of South America. Accordingly, the distribution determined from ¹³⁷Cs inventories in
265 undisturbed soil profiles is mostly valid for Brazil, Uruguay, Argentina and Chile.

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

278 A significant linear relationship was found between ^{137}Cs inventories in reference soil sites and annual
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2 279 rainfall rates provided by the authors ($R^2=0.43$, $n=92$, $p < 0.001$). However, a wide dispersion of ^{137}Cs
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4 280 inventories values is found for similar levels of precipitation (Fig. 4). In contrast, when reorganising the
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7 281 data according to the climatic circulation cells in which they are located, a significant relationship is
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9 282 observed between mean annual rainfall rates and ^{137}Cs inventories in soils of Argentina and Chile
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11 283 ($R^2=0.72$, $n=54$, $p < 0.001$). Furthermore, another linear relationship, although less significant, is found
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13 284 for those reference soil sites located between the Equator and mid-latitudes ($R^2=0.39$, $n=38$, $p < 0.001$).
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17 285 Contrary to what has been observed for precipitation, no significant relationship was found between
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19 286 the altitude reported by the authors at the reference sites and the ^{137}Cs inventories ($n=74$). However,
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22 287 when the analysis is performed for ^{137}Cs inventories in reference soil sites located at the same latitude
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24 288 although at a different altitude, it can be observed that ^{137}Cs inventories tends to increase with the
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26 289 altitude (data not shown). This observation confirms that made by Handl et al. (2008) who stated that
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29 290 maximum values were observed in regions of high altitude between 23° and 29° South latitude. In Chile,
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31 291 Schuller et al. (2002) found a similar relationship for reference soil sites located in the central part of
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33 292 the country, with higher values in elevated altitude areas, which are also exposed to higher rainfall.
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36 293 Furthermore, a low correlation is observed between the altitude of the sampling sites and the ^{137}Cs
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38 294 inventories in reference sites located between the mid-latitudes and Southern Chile ($R^2 = 0.20$, $n=52$,
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41 295 $p < 0.01$). A step wise multiple regression indicated a significant relationship between observed and
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43 296 predicted ^{137}Cs inventories with a model including geographical information and annual rainfall rates
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45 297 ($R^2 = 0.74$, $n = 71$, $p < 0.001$) (Fig. 5).

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50 51 52 299 **3. Mapping ^{137}Cs initial fallout in South American soils**

53 300 54 301 55 56 302 **3.1. Context**

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305 To the best of our knowledge, only very few studies (n= 7) were conducted to map bomb-derived ¹³⁷Cs
306 fallout (Table 2). The spatial distribution of ¹³⁷Cs deposition was assessed when results of global fallout
307 long-term monitoring programmes were available during the nuclear weapon tests period. By means
308 of a GIS-based approach, Wright et al. (1999) used ¹³⁷Cs deposition data from the Arctic Monitoring and
309 Assessment Programme (AMAP) with annual rainfall rates for the period 1955-1985 to predict ¹³⁷Cs
310 deposition at the scale of the Arctic. Similarly, Pálsson et al. (2006) used activity concentrations of
311 global fallout in precipitation at Rjúpnahæð, in addition to ¹³⁷Cs measurements in undisturbed soil
312 profiles, to predict the spatial variation in global fallout of ¹³⁷Cs in Iceland. In Australia, Chappell et al.
313 (2011a) used indicator co-simulation between 141 inventories in reference soil sites and gridded mean
314 annual rainfall (1954–1990) to create a baseline map of ¹³⁷Cs fallout for Australian soils. More recently,
315 Meusburger et al. (2020) used a digital soil mapping approach (McBratney et al., 2003) to predict the
316 proportions and sources of artificial radionuclides in soils of several countries of Western Europe
317 (France, Belgium, Switzerland, Southern Germany and Northern Italy). Their approach used a
318 generalized additive models (GAM) with environmental factors.

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320 3.2. Digital mapping of ¹³⁷Cs fallout in soils of South America

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

331 resolution of 30-arc seconds, calculated from the 1950-2000 period (Hijmans et al., 2005), and the
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2 332 spatial coordinates (X and Y in meters).
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5 333 Based on a cross validation procedure, a partial least square regression (PLSR) approach was selected.
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7 334 The theory underlying PLSR has been described in several statistical textbooks and articles
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10 335 (Höskuldsson, 1988; Tenenhaus, 1998). PLSR could be deemed as a generalization of the multiple linear
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12 336 regression (Gerlach et al., 1979). PLSR is of particular interest because, unlike multiple linear
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15 337 regression, it can analyze noisy data with numerous collinear variables.
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18 338 The statistical analyses as well as the DSM procedures described in this section were carried out with
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20 339 the R software (Team R Core, 2013) and the following packages: caret (Kuhn, 2008), sf (Pebesma,
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22 340 2018), ithir (Malone, 2015), ggplot2 (Wickham, 2016), raster (Hijmans et al., 2015), corrplot (Wei et al.,
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25 341 2017) and clhs (Roudier, 2011).
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30 343 Figure 6 shows the correlogram plot and the associated coefficients of correlation. The highest
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32 344 correlation was found between ^{137}Cs inventories and average monthly rainfall in August and
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35 345 September ($r=0.7$, $p < 0.001$), followed by average monthly rainfall in May, June, July and average
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37 346 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
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40 347 correlation was observed for elevation. Based on this preliminary statistical analysis, seven covariates
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42 348 (latitude, longitude, average monthly rainfall in June, July, August, September and mean annual rainfall
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44 349 rates) were selected to perform the PLSR.
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48 350 Following the cross validation procedure, we showed that our quantitative model explained 46% of
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50 351 the ^{137}Cs variability observed and tended to underestimate inventories $> 800 \text{ Bq m}^{-2}$. The predicted
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52 352 ^{137}Cs inventories followed a right-skewed statistical distribution, with an average of 348 Bq m^{-2} and a
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54 353 standard deviation of 237 Bq m^{-2} . Highest values (between 1501 to 2057 Bq m^{-2}) were found in
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57 354 Colombia and few areas of the Andean Cordillera, while the vast majority of ^{137}Cs inventories lower
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59 355 than 100 Bq m^{-2} were found in the North-Eastern part of Brazil (Fig. 7). Surprisingly, high ^{137}Cs
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1 356 inventories are predicted for countries located near the equator, where fallout should be the lowest
2 357 according to UNSCEAR (2000). Of note, the highest inventories are mainly located along the Andean
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4 358 Cordillera at high altitudes. Taking into account that the model does not include elevation as a
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7 359 covariable, the deposition pattern of ^{137}Cs is closely linked to that of rainfall rates and it may reflect
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9 360 the orographic effects that occur in mountainous environments. However, these mountainous areas
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11 361 predominantly consist of rock outcrops and bare soil surfaces. Consequently, areas above 1,800 m a.s.l
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13 362 have been masked using the GMTED2010 30-arc-second elevation database (Danielson and Gesch,
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15 363 2011). As previously mentioned, most of the ^{137}Cs inventories in undisturbed soil profiles used to
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17 364 develop the digital soil mapping approach were measured in the eastern part of South America and in
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19 365 Southern Chile. The estimates provided for the other regions (i.e. northern and western parts of the
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21 366 subcontinent) should therefore be interpreted with caution. Additional sampling in these areas will be
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23 367 necessary to improve model predictions of ^{137}Cs global fallout in South America.
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29 368 The baseline map of ^{137}Cs inventories in reference soil sites (Bq m^{-2} , 2020) with a spatial resolution of
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31 369 2 km (projection: WGS 84 - World Geodetic System 1984) was compiled using ESRI ArcGIS 10.6
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33 370 Desktop. The validity of the proposed approach to predict the spatial distribution of the reference
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35 371 levels of fallout ^{137}Cs at the subcontinental-scale of South America relies on the assumption that studies
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37 372 listed in our database followed the recommendations to select and sample undisturbed soil sites. Since
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39 373 the coefficient of variation of multiple samples collected to characterize one reference site is not
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41 374 always reported by authors, the observations uncertainties are difficult to quantify. As reported in the
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43 375 literature, ^{137}Cs inventories in reference soil sites have a coefficient of variation of approximately 20%,
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45 376 as a consequence of random and systematic spatial variability, sampling variability and measurement
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47 377 precisions (Loughran et al., 2002; Owens and Walling, 1996; Pennock, 2000). Accordingly, the map
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49 378 predictions provided in this study for each grid should be interpreted as a trend rather than a single
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51 379 accurate value.
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381 A non-parametric bootstrap approach was used to quantify the prediction uncertainties of the map
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2 382 (Efron and Tibshirani, 1994; Liddicoat et al., 2015; Rossel et al., 2015). In this approach, data used for
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4 383 model calibration was selected using random sampling with replacement, with sample size equal to 95
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7 384 % of the number of data in the available dataset. This calibration step is iterated 100 times leading to
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9 385 100 contributing predictions at each prediction location. These predictions collectively constituted an
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11 386 empirical probability distribution (EDP) of the ^{137}Cs inventories. We computed the 95 % prediction
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13 387 confidence interval by subtracting the 97.5% quantile to the 2.5 % quantile of the EDP. For clarity, the
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16 388 coefficient of variation (in %) was computed by dividing the average prediction by the 95% confidence
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18 389 interval (Fig. 8). For most part of South America, the relative error was comprised between 0 and 10
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21 390 %. The highest uncertainties could be observed in two areas located in the North-Eastern part of Brazil
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23 391 and along the Pacific coast of Peru, where the map predictions of ^{137}Cs inventories in reference soil
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26 392 sites are the lowest (0 to 100 Bq m⁻² in 2020).

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31 395 3.3. Improving spatial predictions through additional sampling

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37 397 Sampling design plays an essential role in a digital soil mapping approach as it controls the estimation
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39 398 of the statistical model parameters and also the spatial predictions. A sound sampling design is
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42 399 expected to provide a scheme of representative samples covering the study area with a relatively small
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44 400 sample size for financial and logistical reasons. If it is assumed that the soil property is linked to
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46 401 environmental covariates, a robust strategy is to ensure that the measurements are also uniformly
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49 402 spread in the feature (i.e. covariates) space. However, the collected data of the present study did not
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51 403 follow any sampling design as they were compiled from various independent studies, leading to large
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53 404 areas devoid of sampling observations or places with clusters. Accordingly, an experiment was
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56 405 conducted to generate new sampling locations in order to fill the gap in feature space of the selected
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58 406 environmental variables. This can be achieved using conditioned Latin Hypercube sampling (cLHS)

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

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

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

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

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434 In Argentina, ¹³⁷Cs soil profiles have been collected in the Pampa Ondulada region of Buenos Aires
435 Province (Bujan et al., 2003), in soils of La Plata region (Montes et al., 2013), in natural and semi-natural
436 grassland areas of San Luis Province (Ayub et al., 2008) and in the Patagonian Andean forests (La
437 Manna et al., 2019). These studies showed that ¹³⁷Cs inventories were sufficient in these regions to
438 calculate soil redistribution rates based on this technique. Of note, there is a lack of ¹³⁷Cs inventory
439 data in the literature for the Argentinian provinces located between 25 to 40° South latitudes, where
440 inventories in reference soil sites should be the highest of South America according to UNSCEAR
441 (2000). Thus, the ¹³⁷Cs technique should be successfully implemented in this country exposed to
442 elevated levels of both water and wind erosion (Ares et al., 2016; Mendez and Buschiazzo, 2010).

443 In Uruguay, only two ¹³⁷Cs inventory had been documented so far in the literature (Alonso et al., 2012;
444 Tassano et al., 2020). Alonso et al. (2012) assessed soil erosion rates in a forested micro-catchment
445 occupied by eucalyptus plantations within the Río Negro River basin. The three additional ¹³⁷Cs
446 inventories collected in Uruguay by co-authors of the current research remained in the same order of
447 magnitude, with an average value of $336 \pm 13 \text{ Bq m}^{-2}$. These values are consistent with ¹³⁷Cs inventories
448 found in the neighbouring Rio Grande do Sul state, in southernmost Brazil, where ¹³⁷Cs inventories
449 followed an increasing gradient from the Uruguayan border in the South ($315 \pm 22 \text{ Bq m}^{-2}$) to the North
450 ($1022 \pm 292 \text{ Bq m}^{-2}$) (Didone et al., 2019; Handl et al., 2008; Minella et al., 2014).

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

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2 458 Despite the high mean annual rainfall rates observed in the equatorial regions, low ^{137}Cs inventories in
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4 459 reference soil sites were observed in the range of latitudes comprised between 2°N and 10°S. As
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6 460 previously mentioned, one reason that may explain this observation is the upward circulation of air
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8 461 masses as a consequence of Hadley circulation in this latitude band. Soil erosion studies based on ^{137}Cs
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10 462 inventories should be difficult to conduct in the North-Eastern part of Brazil. Furthermore, to the best
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12 463 of our knowledge, no studies on ^{137}Cs inventories in reference soil sites have been conducted in
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14 464 Suriname, Guyana, Venezuela, Colombia and Peru.

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17 465 One study investigating soil erosion in a mountainous watershed of Ecuador (2°S) with the ^{137}Cs
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19 466 technique was not retained in the current review because geographical information of ^{137}Cs inventories
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21 467 in reference soil site was erroneous (Henry et al., 2013). In this study, no flat undisturbed sites that can
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23 468 serve as typical reference sites were found, and reference sites were selected based on ^{137}Cs activity
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25 469 patterns with depth and land use history. A mean ^{137}Cs reference value of $2260 \pm 330 \text{ Bq m}^{-2}$ was
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27 470 established with no decay-correction date presented. Although this value should be taken with
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29 471 caution, it appears valuable to investigate ^{137}Cs initial fallout in equatorial regions of South America.
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31 472 Indeed, anomalous high values of ^{137}Cs activities were also observed in surface soils of Venezuela
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33 473 (LaBrecque et al., 2001) and one ^{137}Cs inventory in a reference soil site of French Guiana (de Tombeur
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35 474 et al., 2020) was found to be twice than what was expected from the UNSCEAR predictions for this
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37 475 latitude band (1022 ± 293 compared to $509 \pm 57 \text{ Bq m}^{-2}$, decay-corrected to 2016).
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44 45 46 477 **5. Perspectives for future research**

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52 480 Beside the fact that baseline maps of ^{137}Cs fallout are of fundamental importance in case of future
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54 481 accidental radionuclide emissions, their use in geomorphological studies provides an opportunity to
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56 482 reconstruct soil redistribution due to soil erosion processes at larger scales (Chappell et al., 2011b;
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58 483 Meusburger et al., 2020). Although estimates of erosion rates at continental scales are debated (Fiener
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484 and Auerswald, 2016; Panagos et al., 2015; Panagos et al., 2016), prediction of ^{137}Cs initial fallout at
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2 485 kilometric scales offers the potential to increase our knowledge of soil erosion processes in catchments
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4 486 through the use of the ^{137}Cs inventory technique and its upscaling (Lizaga et al., 2018). In general, the
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7 487 spatial distribution of anthropogenic radionuclides in soils and their use as tracers of environmental
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9 488 processes is of significant importance for Earth and atmospheric sciences (Bhandari, 1970; Everett et
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12 489 al., 2008; Hirose, 2012; Igarashi et al., 2011; Jagercikova et al., 2017).

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16 491 Considering that no radioactive fallout occurred since 1980 in South America, and as a consequence of
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19 492 radioactive decay, ^{137}Cs activities in soils of South America will continue decreasing and become
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21 493 increasingly difficult to measure without ultra-low background gamma spectrometry facilities (Evrard
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23 494 et al., 2020). The development of surrogate tracers appears necessary for further geomorphological
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26 495 studies using fallout radionuclides, especially in the Southern hemisphere that received only 23% of
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28 496 the total bomb-fallout emitted worldwide according to UNSCEAR (2000). In the last several decades,
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31 497 the efficiency of the $^{239+240}\text{Pu}$ inventory technique to quantify soil erosion rates has been demonstrated
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33 498 in countries located in the Northern hemisphere (Alewell et al., 2014; Alewell et al., 2017; Meusbürger
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35 499 et al., 2016). To the best of our knowledge, one study investigating soil erosion rates using ^{239}Pu was
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38 500 carried out in Australia (Lal et al., 2020). In contrast to ^{137}Cs and $^{239+240}\text{Pu}$, the continuous input of fallout
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40 501 radionuclides including excess lead-210 ($^{210}\text{Pb}_{\text{xs}}$) should be considered as an efficient tracer of soil
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42 502 erosion, especially in regions where low ^{137}Cs fallout occurred (Evrard et al., 2020; Gaspar et al., 2013;
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45 503 Porto et al., 2009; Walling et al., 2011). The effectiveness of these techniques in South America remains
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47 504 unknown and it should be investigated to assess soil redistribution under climate and land use changes.
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53 506 Overall, a significant lack of information about ^{137}Cs inventories in reference soil sites is observed in
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56 507 areas of South America exposed to extensive clearcutting, overgrazing and cropping intensification. As
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58 508 an example, central Brazil in general and the Cerrado Biome in particular (Fig. 9) should have received
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509 sufficient levels of ^{137}Cs fallout to investigate soil redistribution rates induced by deforestation and
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2 510 land uses changes. A better characterization of ^{137}Cs fallout at the scale of South America is also
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4 511 required with the addition of soil inventory measurements in North-Western parts of South America,
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7 512 where some areas should have received sufficient fallout to conduct geomorphological studies despite
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9 513 what was previously expected from the UNSCEAR reports. Accordingly, future research should strive
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11 514 to ensure that basic information (e.g. rainfall databases used) and details on the sampling design (e.g.
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13 515 number of soil profiles used to estimate ^{137}Cs inventories in reference soil sites) are properly
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15 516 documented to improve future model predictions and better consider the issues of uncertainty and
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18 517 data reliability.
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24 519 **6. Conclusions**

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29 522 Based on a compilation of published information, additional measurements and rainfall data over the
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31 523 period 1950-2000, this work represents one of the first approaches to spatialize the reference levels
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33 524 of fallout ^{137}Cs at the subcontinental-scale of South America. The current research demonstrates that
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35 525 the ^{137}Cs inventories technique should be appropriate to assess soil redistribution rates during the
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37 526 agricultural intensification period in Chile, Argentina, Uruguay and Southern Brazil where detectable
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39 527 levels of ^{137}Cs can be expected to be found in sites exposed to erosion. This technique should
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41 528 theoretically be applicable in other countries where no information was available to date, such as
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43 529 Paraguay, Bolivia and Peru. Further investigations should be conducted in equatorial regions where
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45 530 information on ^{137}Cs fallout is scarce. Our results indicate that ^{137}Cs inventories in this region may be
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47 531 higher than expected from the UNSCEAR reports. Additional sampling is necessary to verify whether it
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49 532 will be either complicated or impossible to quantify soil erosion using bomb-derived ^{137}Cs in equatorial
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51 533 regions exposed to extensive clearcutting and agricultural expansion.
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58 534 In addition to the priority complementary sampling locations identified through the use of conditioned
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535 Latin Hypercube sampling (cLHS), the South American continent could be subdivided into regions
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2 536 corresponding to different biomes in which the fate of the global ¹³⁷Cs fallout is expected to be
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4 537 homogeneous. Consequently, future sampling campaigns should strive to ensure that all biomes are
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7 538 well covered with a sufficient number of soil profiles.

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9 539 The map generated can be used both to validate ¹³⁷Cs inventories collected in the field or as a decision-
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11 540 support tool to guide the implementation of the ¹³⁷Cs technique in intensive agricultural landscapes of
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13 541 South America. This baseline map will also be particularly useful for a wide range of Earth science
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16 542 applications, including the vertical transfers in soils, the circulation of air masses and ocean currents.
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18 543 Of note, this map is provisional as it is only based on the data published until early 2020 and it can be
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21 544 optimized through the incorporation of additional ¹³⁷Cs inventory measurements in South America.
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43
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45 46 47 554 **7. References**

48 555
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50
51 556 Alewell, C., Meusburger, K., Juretzko, G., Mabit, L. and Ketterer, M.E., 2014. Suitability of ²³⁹⁺²⁴⁰Pu
52 557 and ¹³⁷Cs as tracers for soil erosion assessment in mountain grasslands. *Chemosphere*, 103:
53 558 274-280.
54 559 Alewell, C., Pitois, A., Meusburger, K., Ketterer, M. and Mabit, L., 2017. ²³⁹⁺²⁴⁰Pu from
55 560 “contaminant” to soil erosion tracer: Where do we stand? *Earth-Science Reviews*, 172: 107-
56 561 123.
57 562 Almgren, S., Nilsson, E., Erlandsson, B. and Isaksson, M., 2006. GIS supported calculations of ¹³⁷Cs
58 563 deposition in Sweden based on precipitation data. *Science of the Total Environment*, 368(2-
59 564 3): 804-813.
60
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565 Alonso, J., Audicio, P., Martinez, L., Scavone, M. and Rezzano, E., 2012. Comparison of measured
1 566 ¹³⁷Cs data and USLE/RUSLE simulated long-term erosion rates. *Agrociencia-Sitio en*
2 567 *Reparación*, 16(3): 261-267.

3 568 Aoyama, M., Hirose, K. and Igarashi, Y., 2006. Re-construction and updating our understanding on
4 569 the global weapons tests ¹³⁷Cs fallout. *Journal of Environmental Monitoring*, 8(4): 431.

5 570 Ares, M.G., Bongiorno, F., Holzman, M., Chagas, C., Varni, M. and Entraigas, I., 2016. Water erosion
6 571 and connectivity analysis during a year with high precipitations in a watershed of Argentina.
7 572 *Hydrology Research*, 47(6): 1239-1252.

8 573 Ayub, J.J., Velasco, R., Rizzotto, M., Quintana, E. and Aguiar, J., 2008. ⁴⁰K, ¹¹⁵Cs and ²²⁶Ra Soil and
9 574 Plant Content in Seminatural Grasslands of Central Argentina, AIP Conference Proceedings.
10 575 AIP, pp. 273-276.

11 576 Bacchi, O.O.S., Reichard, K., Sparovek, G. and Ranieri, S.B.L., 2000. Soil erosion evaluation in a small
12 577 watershed in Brazil through ¹³⁷Cs fallout redistribution analysis and conventional models.
13 578 *Acta Geologica Hispanica*, 35(3-4): 251-259.

14 579 Bakker, M.M., Govers, G., Jones, R.A. and Rounsevell, M.D.A., 2007. The Effect of Soil Erosion on
15 580 Europe's Crop Yields. *Ecosystems*, 10(7): 1209-1219.

16 581 Bakker, M.M., Govers, G. and Rounsevell, M.D., 2004. The crop productivity–erosion relationship: an
17 582 analysis based on experimental work. *Catena*, 57(1): 55-76.

18 583 Banfield, C.C., Braun, A.C., Barra, R., Castillo, A. and Vogt, J., 2018. Erosion proxies in an exotic tree
19 584 plantation question the appropriate land use in Central Chile. *CATENA*, 161: 77-84.

20 585 Becker, A.G., Moraes, B.S., Menezes, C.C., Loro, V.L., Santos, D.R., Reichert, J.M. and Baldisserotto, B.,
21 586 2009. Pesticide contamination of water alters the metabolism of juvenile silver catfish,
22 587 *Rhamdia quelen*. *Ecotoxicol Environ Saf*, 72(6): 1734-9.

23 588 Bennett, B.G., 2002. Worldwide dispersion and deposition of radionuclides produced in atmospheric
24 589 tests : Fallout from atmospheric nuclear tests : Impact on science and society. *Health Physics*,
25 590 82(5): 644-655.

26 591 Bhandari, N., 1970. Transport of air in the stratosphere as revealed by radioactive tracers. *Journal of*
27 592 *Geophysical Research*, 75(15): 2927-2930.

28 593 Bouisset, P., Nohl, M., Bouville, A. and Leclerc, G., 2018. Inventory and vertical distribution of ¹³⁷Cs,
29 594 ²³⁹⁺²⁴⁰Pu and ²³⁸Pu in soil from Raivavae and Hiva Oa, two French Polynesian islands in the
30 595 southern hemisphere. *Journal of Environmental Radioactivity*, 183: 82-93.

31 596 Buesseler, K.O. and Benitez, C.R., 1994. Determination of mass accumulation rates and sediment
32 597 radionuclide inventories in the deep Black Sea. *Deep-Sea Research Part I-Oceanographic*
33 598 *Research Papers*, 41(11-12): 1605-1615.

34 599 Bujan, A., Santanatoglia, O.J., Chagas, C., Massobrio, M., Castiglioni, M., Yanez, M., Ciallella, H. and
35 600 Fernandez, J., 2003. Soil erosion evaluation in a small basin through the use of Cs-137
36 601 technique. *Soil & Tillage Research*, 69(1-2): 127-137.

37 602 Camargo, F.A.O., Silva, L.S., Merten, G.H., Carlos, F.S., Baveye, P.C. and Triplett, E.W., 2017. Brazilian
38 603 Agriculture in Perspective. *Advances in Agronomy*, pp. 53-114.

39 604 Cambray, R., Playford, K., Lewis, G. and Carpenter, R., 1989. Radioactive fallout in air and rain: results
40 605 to the end of 1988. AERE R13575. Atomic Energy Authority, London, UK.

41 606 Chappell, A., Hancock, G., Viscarra Rossel, R.A. and Loughran, R., 2011a. Spatial uncertainty of
42 607 the ¹³⁷Cs reference inventory for Australian soil. *Journal of Geophysical Research*, 116(F4).

43 608 Chappell, A., Viscarra Rossel, R.A. and Loughran, R., 2011b. Spatial uncertainty of ¹³⁷Cs-derived net
44 609 (1950s–1990) soil redistribution for Australia. *Journal of Geophysical Research: Earth Surface*,
45 610 116(F4).

46 611 Chartin, C., Evrard, O., Salvador-Blanes, S., Hirschberger, F., Van Oost, K., Lefèvre, I., Daroussin, J. and
47 612 Macaire, J.-J., 2013. Quantifying and modelling the impact of land consolidation and field
48 613 borders on soil redistribution in agricultural landscapes (1954–2009). *Catena*, 110: 184-195.

49 614 Correchel, V., Bacchi, O.O.S., Reichardt, K. and De Maria, I.C., 2005. Random and systematic spatial
50 615 variability of ¹³⁷Cs inventories at reference sites in South-Central Brazil. *Scientia Agricola*,
51 616 62(2): 173-178.

617 de Tombeur, F., Cornu, S., Bourlès, D., Duvivier, A., Pupier, J., Aster, T., Brossard, M. and Evrard, O.,
1 618 2020. Retention of ^{10}Be , ^{137}Cs and ^{210}Pb s in soils: Impact of physico-chemical
2 619 characteristics. *Geoderma*, 367.

3 620 Didone, E.J., Minella, J.P.G., Schneider, F.J.A., Londero, A.L., Lefevre, I. and Evrard, O., 2019.
4 621 Quantifying the impact of no-tillage on soil redistribution in a cultivated catchment of
5 622 Southern Brazil (1964-2016) with Cs-137 inventory measurements. *Agriculture Ecosystems &*
6 623 *Environment*, 284.

8 624 Efron, B. and Tibshirani, R.J., 1994. An introduction to the bootstrap. CRC press.

9 625 Ehhalt, D., 1973. Turnover times of ^{137}Cs and HTO in the troposphere and removal rates of natural
10 626 aerosol particles and water vapor. *Journal of Geophysical Research*, 78(30): 7076-7086.

11 627 Everett, S., Tims, S., Hancock, G., Bartley, R. and Fifield, L.K., 2008. Comparison of Pu and ^{137}Cs as
12 628 tracers of soil and sediment transport in a terrestrial environment. *Journal of Environmental*
13 629 *Radioactivity*, 99(2): 383-393.

15 630 Evrard, O., Biielders, C.L., Vandaele, K. and van Wesemael, B., 2007. Spatial and temporal variation of
16 631 muddy floods in central Belgium, off-site impacts and potential control measures. *Catena*,
17 632 70(3): 443-454.

19 633 Evrard, O., Chaboche, P.-A., Ramon, R., Foucher, A. and Lacey, J.P., 2020. A global review of
20 634 sediment source fingerprinting research incorporating fallout radiocesium (^{137}Cs).
21 635 *Geomorphology*: 107103.

22 636 FAO, 2015. Status of the world's soil resources: main report. FAO : ITPS, Rome.

23 637 FAO/IAEA, 2017. Use of ^{137}Cs for soil erosion assessment. Fulajtar, E., Mabit, L., Renschler, C.S., Lee
24 638 Zhi Yi, A., . In: F.a.A.O.o.t.U. Nations (Editor), Rome, Italy, pp. 64.

26 639 Fiener, P. and Auerswald, K., 2016. Comment on "The new assessment of soil loss by water erosion in
27 640 Europe" by Panagos et al. (*Environmental Science & Policy* 54 (2015) 438–447).
28 641 *Environmental Science & Policy*, 57: 140-142.

29 642 Foucher, A., Salvador-Blanes, S., Evrard, O., Simonneau, A., Chapron, E., Courp, T., Cerdan, O.,
30 643 Lefèvre, I., Adriaensen, H., Lecompte, F. and Desmet, M., 2014. Increase in soil erosion after
31 644 agricultural intensification: Evidence from a lowland basin in France. *Anthropocene*, 7: 30-41.

33 645 Fukuyama, T., Takenaka, C. and Onda, Y., 2005. ^{137}Cs loss via soil erosion from a mountainous
34 646 headwater catchment in central Japan. *Science of the total environment*, 350(1-3): 238-247.

35 647 Furuichi, T. and Wasson, R.J., 2013. Caesium-137 in Southeast Asia: Is there enough left for soil
36 648 erosion and sediment redistribution studies? *Journal of Asian Earth Sciences*, 77: 108-116.

38 649 Garcia Agudo, E., 1998. Global distribution of Cs-137 inputs for soil erosion and sedimentation
39 650 studies, In: *Use of Caesium-137 in the Study of Soil Erosion and Sedimentation*. IAEA TECDOC
40 651 1028., pp. 117-121.

41 652 Garrett, R.D., Koh, I., Lambin, E.F., De Waroux, Y.I.P., Kastens, J.H. and Brown, J., 2018. Intensification
42 653 in agriculture-forest frontiers: Land use responses to development and conservation policies
43 654 in Brazil. *Global Environmental Change*, 53: 233-243.

45 655 Gaspar, L., Navas, A., Walling, D., Machín, J. and Arozamena, J.G., 2013. Using ^{137}Cs and ^{210}Pb to
46 656 assess soil redistribution on slopes at different temporal scales. *Catena*, 102: 46-54.

47 657 Gerlach, R.W., Kowalski, B.R. and Wold, H.O., 1979. Partial least squares path modelling with latent
48 658 variables, WASHINGTON UNIV SEATTLE LAB FOR CHEMOMETRICS.

49 659 Gilbert, E.S., Land, C.E. and Simon, S.L., 2002. Health effects from fallout. *Health Physics*, 82(5): 726-
50 660 735.

52 661 Handl, J., Sachse, R., Jakob, D., Michel, R., Evangelista, H., Gonçalves, A.C. and de Freitas, A.C., 2008.
53 662 Accumulation of ^{137}Cs in Brazilian soils and its transfer to plants under different climatic
54 663 conditions. *Journal of Environmental Radioactivity*, 99(2): 271-287.

55 664 Health and Safety Laboratory, E., New York 10014, 1977. Final tabulation of monthly strontium-90
56 665 fallout data : 1954-1976.

58 666 Henry, A., Mabit, L., Jaramillo, R.E., Cartagena, Y. and Lynch, J.P., 2013. Land use effects on erosion
59 667 and carbon storage of the Rio Chimbo watershed, Ecuador. *Plant and Soil*, 367(1-2): 477-491.

668 Hijmans, R.J., Cameron, S.E., Parra, J.L., Jones, P.G. and Jarvis, A., 2005. Very high resolution
1 669 interpolated climate surfaces for global land areas. *International Journal of Climatology*,
2 670 25(15): 1965-1978.

3 671 Hijmans, R.J., Van Etten, J., Cheng, J., Mattiuzzi, M., Sumner, M., Greenberg, J.A., Lamigueiro, O.P.,
4 672 Bevan, A., Racine, E.B. and Shortridge, A., 2015. Package 'raster'. R package, 734.

5 673 Hirose, K., 2012. Uranium, thorium and anthropogenic radionuclides as atmospheric tracers,
6 674 *Handbook of Environmental Isotope Geochemistry*. Springer, pp. 591-611.

7 675 Höskuldsson, A., 1988. PLS regression methods. *Journal of chemometrics*, 2(3): 211-228.

8 676 Igarashi, Y., Fujiwara, H. and Jugder, D., 2011. Change of the Asian dust source region deduced from
9 677 the composition of anthropogenic radionuclides in surface soil in Mongolia. *Atmospheric
10 678 Chemistry and Physics*, 11(14): 7069.

11 679 Jagercikova, M., Cornu, S., Bourlès, D., Evrard, O., Hatté, C. and Balesdent, J., 2017. Quantification of
12 680 vertical solid matter transfers in soils during pedogenesis by a multi-tracer approach. *Journal
13 681 of Soils and Sediments*, 17(2): 408-422.

14 682 Keesstra, S.D., Bouma, J., Wallinga, J., Tittonell, P., Smith, P., Cerdà, A., Montanarella, L., Quinton,
15 683 J.N., Pachepsky, Y., van der Putten, W.H., Bardgett, R.D., Moolenaar, S., Mol, G., Jansen, B.
16 684 and Fresco, L.O., 2016. The significance of soils and soil science towards realization of the
17 685 United Nations Sustainable Development Goals. *Soil*, 2(2): 111-128.

18 686 Kuhn, M., 2008. Building predictive models in R using the caret package. *Journal of statistical
19 687 software*, 28(5): 1-26.

20 688 La Manna, L., Gaspar, L., Tarabini, M., Quijano, L. and Navas, A., 2019. Cs-137 inventories along a
21 689 climatic gradient in volcanic soils of Patagonia: Potential use for assessing medium term
22 690 erosion processes. *Catena*, 181.

23 691 LaBrecque, J., Rosales, P. and Cordoves, P., 2001. Anomalously high values of cesium-137 in soils on
24 692 the Peninsula de Paraguana (Venezuela). *Journal of Radioanalytical and Nuclear Chemistry*,
25 693 247(3): 563-566.

26 694 Lal, R., 2015. Restoring Soil Quality to Mitigate Soil Degradation. *Sustainability*, 7(5): 5875-5895.

27 695 Lal, R., Fifield, L., Tims, S., Wasson, R. and Howe, D., 2020. A study of soil erosion rates using ²³⁹Pu,
28 696 in the wet-dry tropics of northern Australia. *Journal of environmental radioactivity*, 211:
29 697 106085.

30 698 Le Roux, G., Duffa, C., Vray, F. and Renaud, P., 2010. Deposition of artificial radionuclides from
31 699 atmospheric Nuclear Weapon Tests estimated by soil inventories in French areas low-
32 700 impacted by Chernobyl. *J Environ Radioact*, 101(3): 211-8.

33 701 Liddicoat, C., Maschmedt, D., Clifford, D., Searle, R., Herrmann, T., Macdonald, L.M. and Baldock, J.,
34 702 2015. Predictive mapping of soil organic carbon stocks in South Australia's agricultural zone.
35 703 *Soil Research*, 53(8): 956-973.

36 704 Lizaga, I., Quijano, L., Gaspar, L. and Navas, A., 2018. Estimating soil redistribution patterns with
37 705 ¹³⁷Cs measurements in a Mediterranean mountain catchment affected by land
38 706 abandonment. *Land Degradation & Development*, 29(1): 105-117.

39 707 Loughran, R., Pennock, D. and Walling, D., 2002. Spatial distribution of caesium-137, *Handbook for
40 708 the assessment of soil erosion and sedimentation using environmental radionuclides*.
41 709 Springer, pp. 97-109.

42 710 Mabit, L., Bernard, C., Lee Zhi Yi, A., Fulajtar, E., Dercon, G., Zaman, M., Toloza, A. and Heng, L., 2018.
43 711 Promoting the use of isotopic techniques to combat soil erosion: An overview of the key role
44 712 played by the SWMCN Subprogramme of the Joint FAO/IAEA Division over the last 20 years.
45 713 *Land Degradation & Development*, 29(9): 3077-3091.

46 714 Mabit, L., Bernard, C., Makhlof, M. and Laverdière, M., 2008. Spatial variability of erosion and soil
47 715 organic matter content estimated from ¹³⁷Cs measurements and geostatistics. *Geoderma*,
48 716 145(3-4): 245-251.

49 717 Mabit, L., Meusburger, K., Fulajtar, E. and Alewell, C., 2013. The usefulness of ¹³⁷Cs as a tracer for
50 718 soil erosion assessment: A critical reply to Parsons and Foster (2011). *Earth-Science Reviews*,
51 719 127: 300-307.

- 720 Macêdo, Í.L., 2009. Estudo de modelos em ambiente de geoprocessamento para a previsão de
 1 721 erosão e assoreamento de reservatórios: O caso da bacia do Rio Indaiá – Uhe Três Marias,
 2 722 Mg. 340.
- 3 723 Malakhov, S.G. and Pudovkina, I.B., 1970. Strontium 90 fallout distribution at middle latitudes of the
 4 724 Northern and Southern Hemispheres and its relation to precipitation. *Journal of Geophysical*
 5 725 *Research*, 75(18): 3623-3628.
- 7 726 Malone, B., 2015. *ithir: Soil data and some useful associated functions*. R package version, 1.
- 8 727 Martin, P. and McBride, J.L., 2012. Radionuclide Behaviour and Transport in the Tropical Atmospheric
 9 728 Environment, *Radioactivity in the Environment*. Elsevier, pp. 59-91.
- 10 729 McBratney, A.B., Santos, M.M. and Minasny, B., 2003. On digital soil mapping. *Geoderma*, 117(1-2):
 11 730 3-52.
- 13 731 McCarthy, M., 1997. Nuclear bomb test fallout may cause many US cancers. *The Lancet*, 350(9075):
 14 732 415.
- 15 733 Mendez, M.J. and Buschiazzo, D.E., 2010. Wind erosion risk in agricultural soils under different tillage
 16 734 systems in the semiarid Pampas of Argentina. *Soil & Tillage Research*, 106(2): 311-316.
- 17 735 Meusburger, K., Evrard, O., Alewell, C., Borrelli, P., Cinelli, G., Ketterer, M., Mabit, L., Panagos, P., van
 18 736 Oost, K. and Ballabio, C., 2020. Plutonium aided reconstruction of caesium atmospheric
 20 737 fallout in European topsoils. *Scientific Reports*, 10(1).
- 21 738 Meusburger, K., Mabit, L., Ketterer, M., Park, J.-H., Sandor, T., Porto, P. and Alewell, C., 2016. A multi-
 22 739 radionuclide approach to evaluate the suitability of ²³⁹⁺ ²⁴⁰Pu as soil erosion tracer. *Science*
 23 740 *of the Total Environment*, 566: 1489-1499.
- 25 741 Minasny, B. and McBratney, A.B., 2006. A conditioned Latin hypercube method for sampling in the
 26 742 presence of ancillary information. *Computers & Geosciences*, 32(9): 1378-1388.
- 27 743 Minella, J.P., Walling, D.E. and Merten, G.H., 2014. Establishing a sediment budget for a small
 28 744 agricultural catchment in southern Brazil, to support the development of effective sediment
 29 745 management strategies. *Journal of Hydrology*, 519: 2189-2201.
- 31 746 Montes, M., Silva, L., Sá, C., Runco, J., Taylor, M. and Desimoni, J., 2013. Inventories and
 32 747 concentration profiles of ¹³⁷Cs in undisturbed soils in the northeast of Buenos Aires
 33 748 Province, Argentina. *Journal of environmental radioactivity*, 116: 133-140.
- 34 749 Montgomery, D.R., 2007. Soil erosion and agricultural sustainability. *Proceedings of the National*
 35 750 *Academy of Sciences*, 104(33): 13268-13272.
- 36 751 Navas, A., Machín, J. and Soto, J., 2005. Assessing soil erosion in a Pyrenean mountain catchment
 37 752 using GIS and fallout ¹³⁷Cs. *Agriculture, Ecosystems & Environment*, 105(3): 493-506.
- 39 753 Navas, A., Serrano, E., López-Martínez, J., Gaspar, L. and Lizaga, I., 2018. Interpreting environmental
 40 754 changes from radionuclides and soil characteristics in different landform contexts of
 41 755 Elephant Island (maritime Antarctica). *Land Degradation & Development*, 29(9): 3141-3158.
- 42 756 Owens, P., Batalla, R., Collins, A., Gomez, B., Hicks, D., Horowitz, A., Kondolf, G., Marden, M., Page,
 43 757 M. and Peacock, D., 2005. Fine-grained sediment in river systems: environmental significance
 44 758 and management issues. *River research and applications*, 21(7): 693-717.
- 46 759 Owens, P.N. and Walling, D.E., 1996. Spatial variability of caesium-137 inventories at reference sites:
 47 760 an example from two contrasting sites in England and Zimbabwe. *Applied Radiation and*
 48 761 *Isotopes*, 47(7): 699-707.
- 49 762 Palsson, S.E., Howard, B.J. and Wright, S.M., 2006. Prediction of spatial variation in global fallout of
 50 763 ¹³⁷Cs using precipitation. *Sci Total Environ*, 367(2-3): 745-56.
- 52 764 Panagos, P., Borrelli, P., Poesen, J., Ballabio, C., Lugato, E., Meusburger, K., Montanarella, L. and
 53 765 Alewell, C., 2015. The new assessment of soil loss by water erosion in Europe. *Environmental*
 54 766 *Science & Policy*, 54: 438-447.
- 55 767 Panagos, P., Borrelli, P., Poesen, J., Meusburger, K., Ballabio, C., Lugato, E., Montanarella, L. and
 56 768 Alewell, C., 2016. Reply to the comment on “The new assessment of soil loss by water
 57 769 erosion in Europe” by Fiener & Auerswald. *Environmental Science & Policy*, 57: 143-150.
- 59 770 Parsons, A.J. and Foster, I.D.L., 2011. What can we learn about soil erosion from the use of ¹³⁷Cs?
 60 771 *Earth-Science Reviews*, 108(1-2): 101-113.

- 772 Parsons, A.J. and Foster, I.D.L., 2013. The assumptions of science. *Earth-Science Reviews*, 127: 308-
 1 773 310.
- 2 774 Pebesma, E.J., 2018. Simple features for R: Standardized support for spatial vector data. *R J.*, 10(1):
 3 775 439.
- 4 776 Pennock, D., 2000. Suitability of ¹³⁷Cs redistribution as an indicator of soil quality. *Acta geológica*
 5 777 *hispanica*: 213-217.
- 7 778 Porto, P., Walling, D.E., Callegari, G. and Capra, A., 2009. Using caesium-137 and unsupported lead-
 8 779 ²¹⁰Pb measurements to explore the relationship between sediment mobilisation, sediment
 9 780 delivery and sediment yield for a Calabrian catchment. *Marine and Freshwater research*,
 10 781 60(7): 680-689.
- 11 782 Quinton, J.N., Govers, G., Van Oost, K. and Bardgett, R.D., 2010. The impact of agricultural soil
 12 783 erosion on biogeochemical cycling. *Nature Geoscience*, 3(5): 311-314.
- 14 784 Racoviteanu, A., 2007. GLIMS Glacier Database, Version 1. Analysis IDs 60827-61235, Accessed
 15 785 1/17/2017.
- 16 786 Ritchie, J. and Ritchie, C., 2007. Bibliography of Publications of ¹³⁷Cesium Studies Related to Erosion
 17 787 and Sediment Deposition. Hydrology and Remote Sensing Laboratory, Beltsville, USA.
- 18 788 Ritchie, J.C. and McHenry, J.R., 1990. Application of Radioactive Fallout Cesium-137 for Measuring
 20 789 Soil Erosion and Sediment Accumulation Rates and Patterns: A Review. *Journal of*
 21 790 *Environment Quality*, 19(2): 215.
- 22 791 Rossel, R.V., Chen, C., Grundy, M., Searle, R., Clifford, D. and Campbell, P., 2015. The Australian
 23 792 three-dimensional soil grid: Australia's contribution to the GlobalSoilMap project. *Soil*
 24 793 *Research*, 53(8): 845-864.
- 26 794 Roudier, P., 2011. *clhs*: a R package for conditioned Latin hypercube sampling. R package version 0.5-
 27 795 1.
- 28 796 Schuller, P., Bunzl, K., Voigt, G., Ellies, A. and Castillo, A., 2003a. Global fallout ¹³⁷Cs accumulation
 29 797 and vertical migration in selected soils from South Patagonia. *Journal of Environmental*
 30 798 *Radioactivity*, 71(1): 43-60.
- 32 799 Schuller, P., Ellies, A., Castillo, A. and Salazar, I., 2003b. Use of ¹³⁷Cs to estimate tillage- and water-
 33 800 induced soil redistribution rates on agricultural land under different use and management in
 34 801 central-south Chile. *Soil & Tillage Research*, 69(1-2): 69-83.
- 35 802 Schuller, P., Voigt, G., Handl, J., Ellies, A. and Oliva, L., 2002. Global weapons' fallout ¹³⁷Cs in soils
 36 803 and transfer to vegetation in south-central Chile. *Journal of Environmental Radioactivity*,
 37 804 62(2): 181-193.
- 39 805 Simon, S.L., Bouville, A. and Land, C.E., 2006. Fallout from nuclear weapons tests and cancer risks:
 40 806 exposures 50 years ago still have health implications today that will continue into the future.
 41 807 *American Scientist*, 94(1): 48-57.
- 42 808 Steffen, W., Broadgate, W., Deutsch, L., Gaffney, O. and Ludwig, C., 2015. The trajectory of the
 43 809 Anthropocene: the great acceleration. *The Anthropocene Review*, 2(1): 81-98.
- 45 810 Steinhauser, G., Brandl, A. and Johnson, T.E., 2014. Comparison of the Chernobyl and Fukushima
 46 811 nuclear accidents: a review of the environmental impacts. *Sci Total Environ*, 470-471: 800-17.
- 47 812 Tagami, K., Tsukada, H. and Uchida, S., 2019. Quantifying spatial distribution of ¹³⁷Cs in reference
 48 813 site soil in Asia. *CATENA*, 180: 341-345.
- 49 814 Tamura, T., 1961. Cesium sorption reactions as indicator of clay mineral structures. *Clays and Clay*
 50 815 *Minerals*, 10(1): 389-398.
- 52 816 Tamura, T. and Jacobs, D., 1960. Structural implications in cesium sorption. *Health physics*, 2(4): 391-
 53 817 398.
- 54 818 Tassano, M., Montañez, A., Nuñez, L., Trasante, T., González, J., Irigoyen, J., Cabral, P. and Cabrera,
 55 819 M., 2020. Spatial cross-correlation between physicochemical and microbiological variables at
 56 820 superficial soil with different levels of degradation. *CATENA*: 105000.
- 58 821 Team R Core, 2013. *R: A language and environment for statistical computing*. Vienna, Austria.
- 59 822 Tenenhaus, M., 1998. *La régression PLS: théorie et pratique*. Editions technip.

823 Turney, C.S.M., Palmer, J., Maslin, M.A., Hogg, A., Fogwill, C.J., Southon, J., Fenwick, P., Helle, G.,
1 824 Wilmshurst, J.M., McGlone, M., Bronk Ramsey, C., Thomas, Z., Lipson, M., Beaven, B., Jones,
2 825 R.T., Andrews, O. and Hua, Q., 2018. Global Peak in Atmospheric Radiocarbon Provides a
3 826 Potential Definition for the Onset of the Anthropocene Epoch in 1965. *Scientific Reports*,
4 827 8(1): 3293.

6 828 UNSCEAR, 1962. Report of the United Nations Scientific Committee on the effects of atomic
7 829 radiation. Annex F : Environmental contamination, New York.

8 830 UNSCEAR, 2000. Effects of Ionizing Radiation. United Nations, New York: 453-487.

9 831 UNSCEAR, 2008. Sources and effects of ionizing radiation: United Nations Scientific Committee on the
10 832 Effects of Atomic Radiation: UNSCEAR 2008 report to the General Assembly, with scientific
11 833 annexes. United Nations, New York.

13 834 Vanwalleghem, T., Gómez, J.A., Infante Amate, J., González de Molina, M., Vanderlinden, K., Guzmán,
14 835 G., Laguna, A. and Giráldez, J.V., 2017. Impact of historical land use and soil management
15 836 change on soil erosion and agricultural sustainability during the Anthropocene.
16 837 *Anthropocene*, 17: 13-29.

17 838 Volchok, H., 1965. Distribution of strontium-90 in surface air during 1963. *Nature*, 206(4988): 1031-
18 839 1033.

20 840 Volchok, H., 1966. The global strontium 90 budget. *Journal of Geophysical Research*, 71(6): 1515-
21 841 1518.

22 842 Wadoux, A.M.-C., Brus, D.J. and Heuvelink, G.B., 2019. Sampling design optimization for soil mapping
23 843 with random forest. *Geoderma*, 355: 113913.

25 844 Walling, D. and He, Q., 2000. The global distribution of bomb-derived ¹³⁷Cs reference inventories.
26 845 Final report on IAEA technical contract, 10361: 1-11.

27 846 Walling, D., Zhang, Y. and He, Q., 2011. Models for deriving estimates of erosion and deposition rates
28 847 from fallout radionuclide (caesium-137, excess lead-210, and beryllium-7) measurements
29 848 and the development of user friendly software for model implementation. Impact of soil
30 849 conservation measures on erosion control and soil quality. IAEA-TECDOC-1665: 11-33.

32 850 Walling, D.E. and Quine, T., 1990. Calibration of caesium-137 measurements to provide quantitative
33 851 erosion rate data. *Land Degradation & Development*, 2(3): 161-175.

34 852 Wei, T., Simko, V., Levy, M., Xie, Y., Jin, Y. and Zemla, J., 2017. Package ‘corrplot’. *Statistician*,
35 853 56(316): e24.

36 854 Wickham, H., 2016. *ggplot2: elegant graphics for data analysis*. springer.

38 855 Wingeyer, A.B., Amado, T.J.C., Perez-Bidegain, M., Studdert, G.A., Varela, C.H.P., Garcia, F.O. and
39 856 Karlen, D.L., 2015. Soil Quality Impacts of Current South American Agricultural Practices.
40 857 *Sustainability*, 7(2): 2213-2242.

41 858 Wright, S.M., Howard, B.J., Strand, P., Nylén, T. and Sickel, M.A.K., 1999. Prediction of ¹³⁷Cs
42 859 deposition from atmospheric nuclear weapons tests within the Arctic. *Environmental
43 860 Pollution*, 104(1): 131-143.

45 861 Zapata, F., 2002. Handbook for the assessment of soil erosion and sedimentation using
46 862 environmental radionuclides, 219. Springer.

47 863 Zapata, F., 2003. The use of environmental radionuclides as tracers in soil erosion and sedimentation
48 864 investigations: recent advances and future developments. *Soil and Tillage Research*, 69(1-2):
49 865 3-13.

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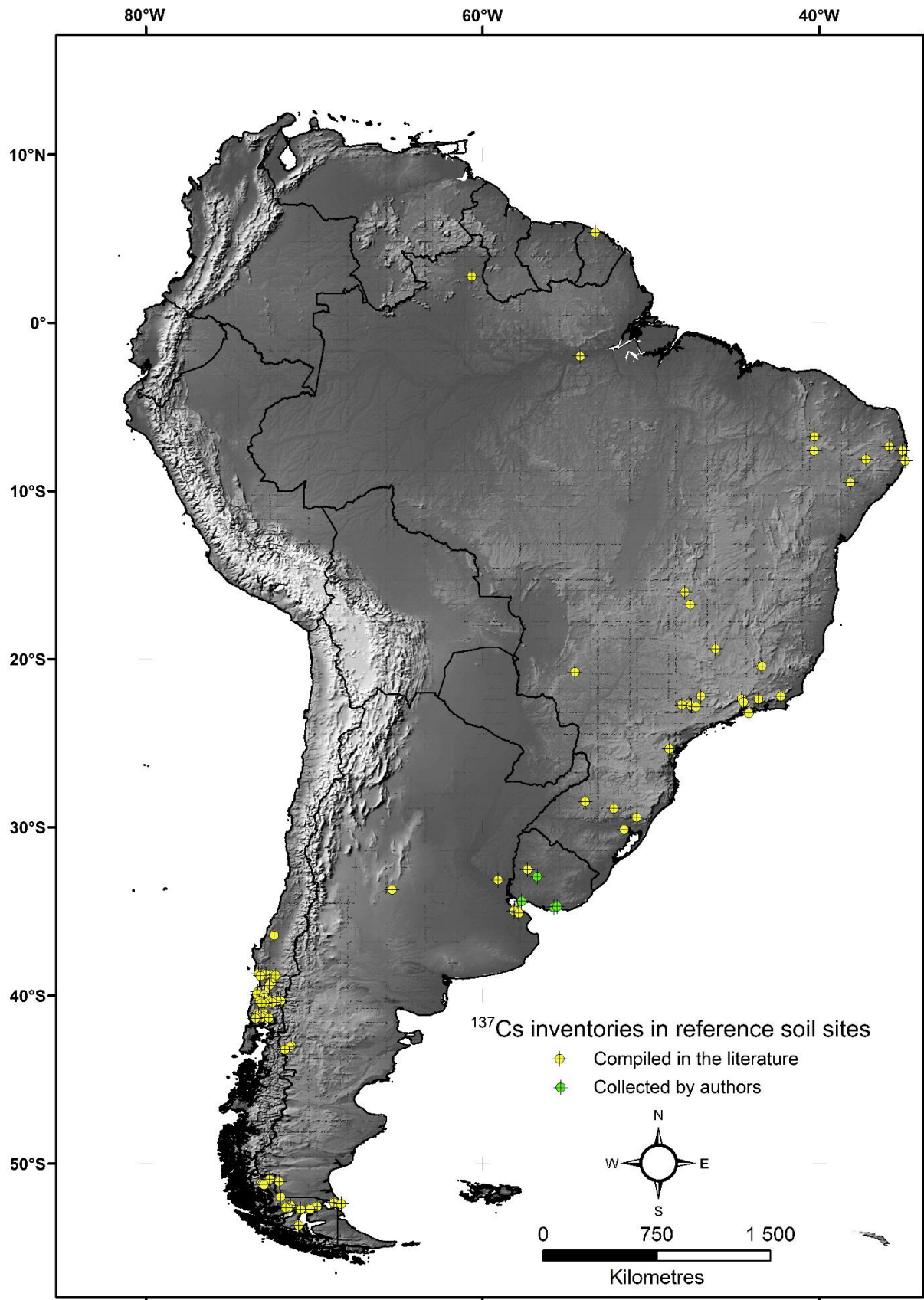


Figure 1: Spatial distribution of ^{137}Cs reference soil sites in undisturbed soil profiles of South America as documented in the literature and collected by the authors.

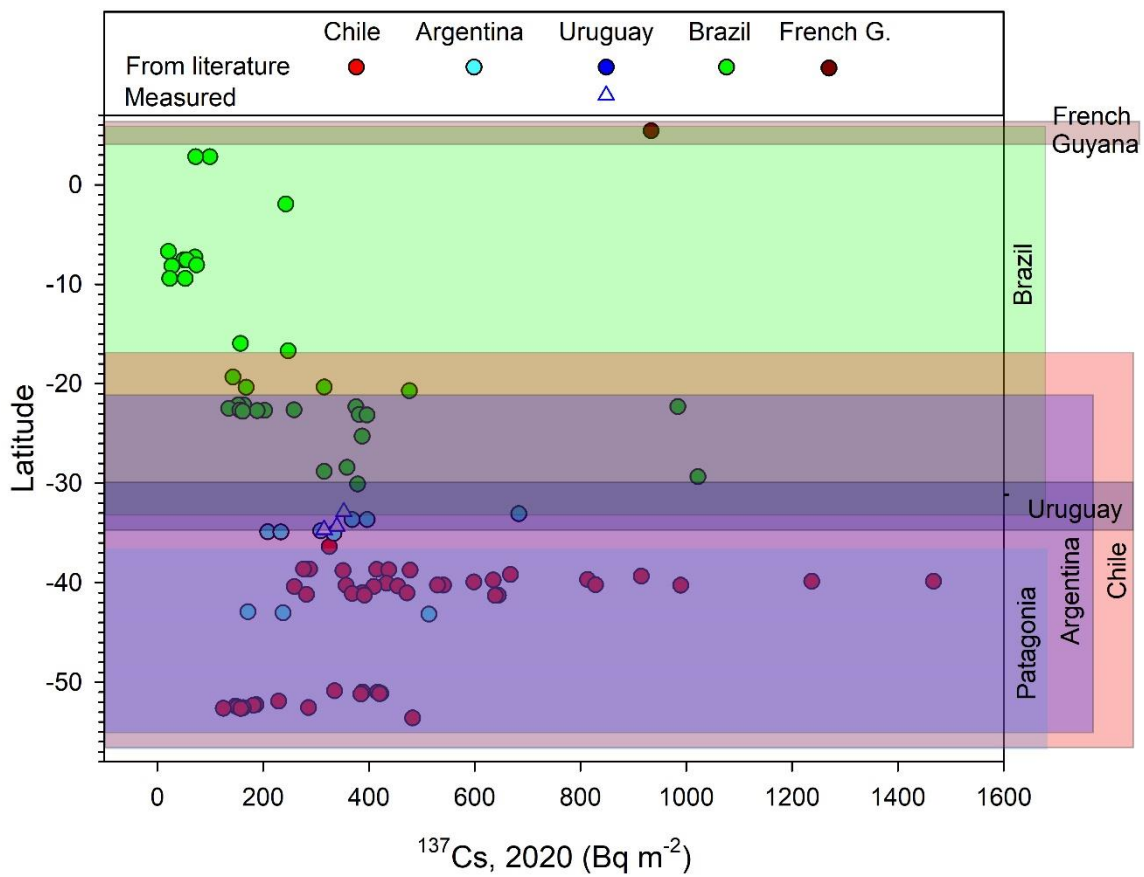


Figure 2: Variation of ^{137}Cs inventories in reference soil sites (Bq m^{-2}), decay-corrected to 2020 and plotted against south latitude.

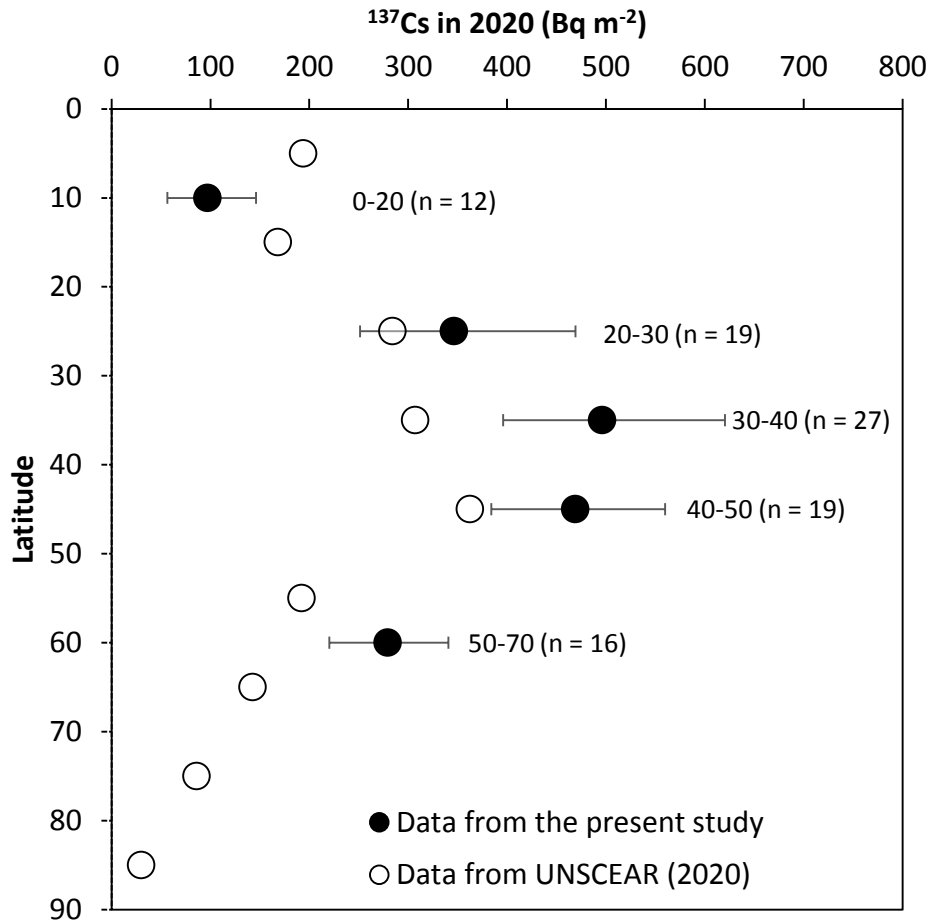


Figure 3: Distribution of mean ^{137}Cs in reference soil sites documented in our review for each 10° latitude bands compared with the distribution of radionuclide fallout from thermonuclear bomb testing with latitude, after UNSCEAR (2000). This distribution was calculated with 5000 samples generated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

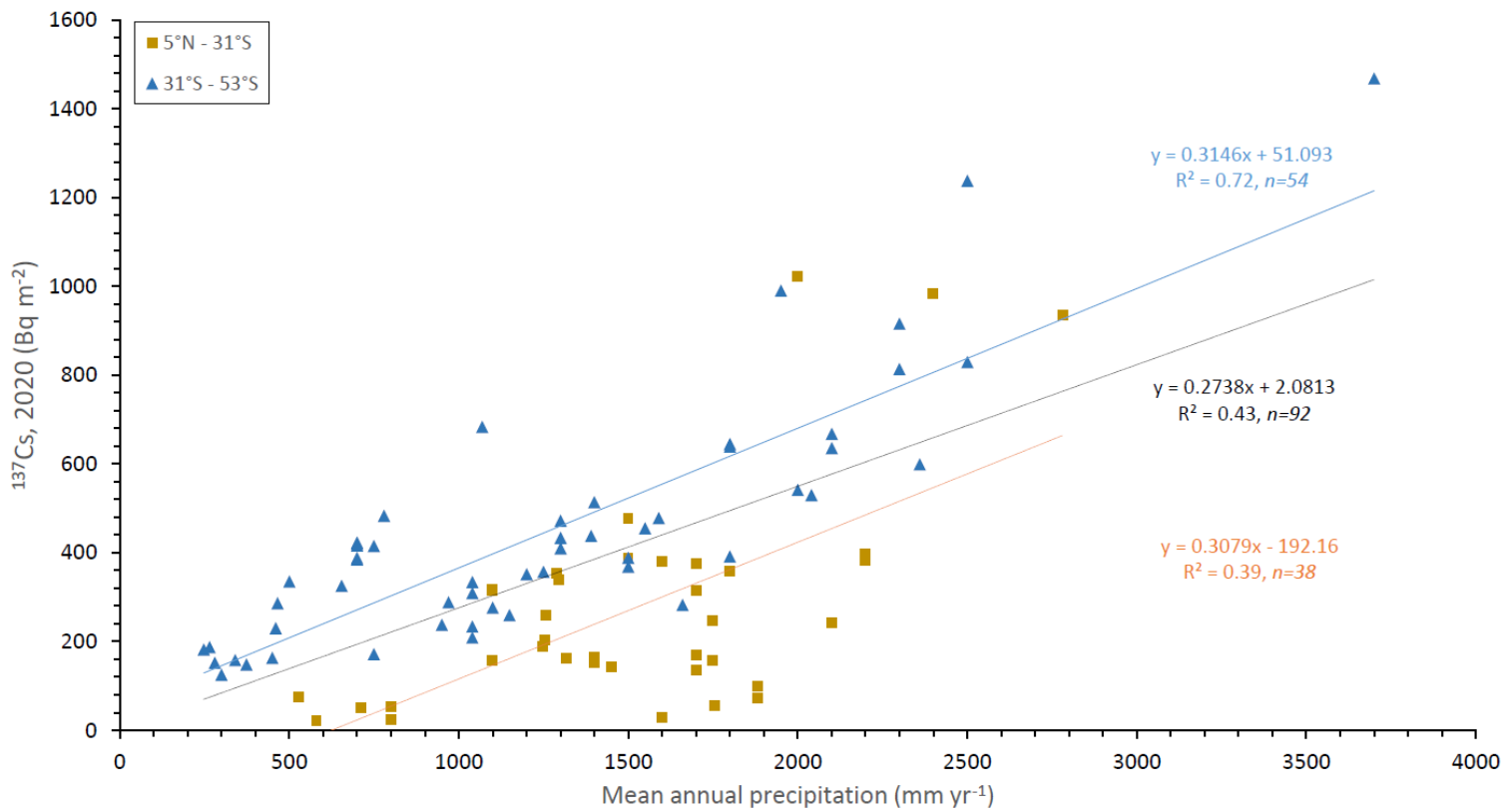


Figure 4: Relationships between mean annual precipitation and ^{137}Cs inventories in reference soil sites for different latitude ranges.

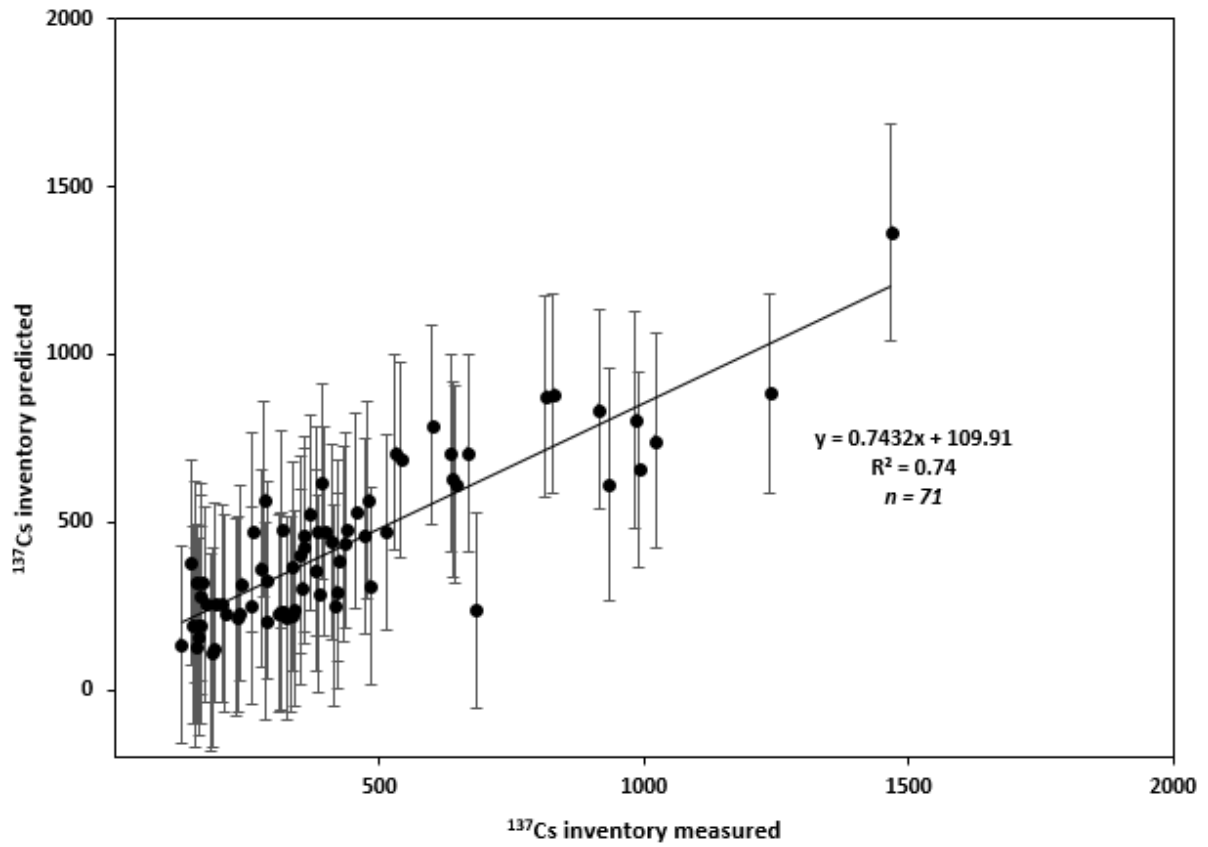


Figure 5: Results of the multiple regressions using latitude, longitude, elevation and rainfall plotted with the 95% confidence intervals.

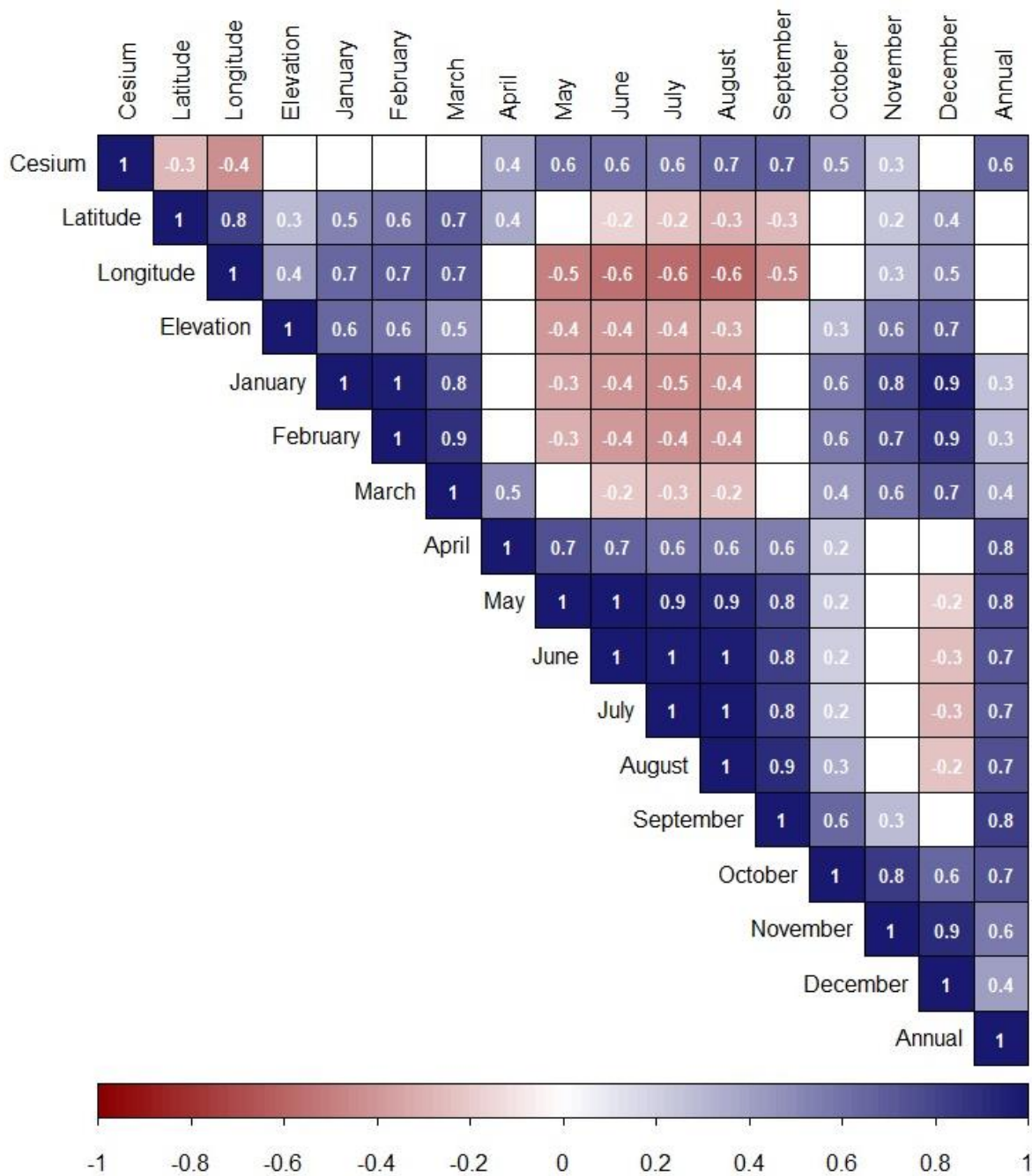


Figure 6: Correlogram plot for ¹³⁷Cs inventories in reference soil sites, mean monthly/annual precipitation rates (1950-2000) and geographical information. Significant levels > 0.1 are represented as white squares.

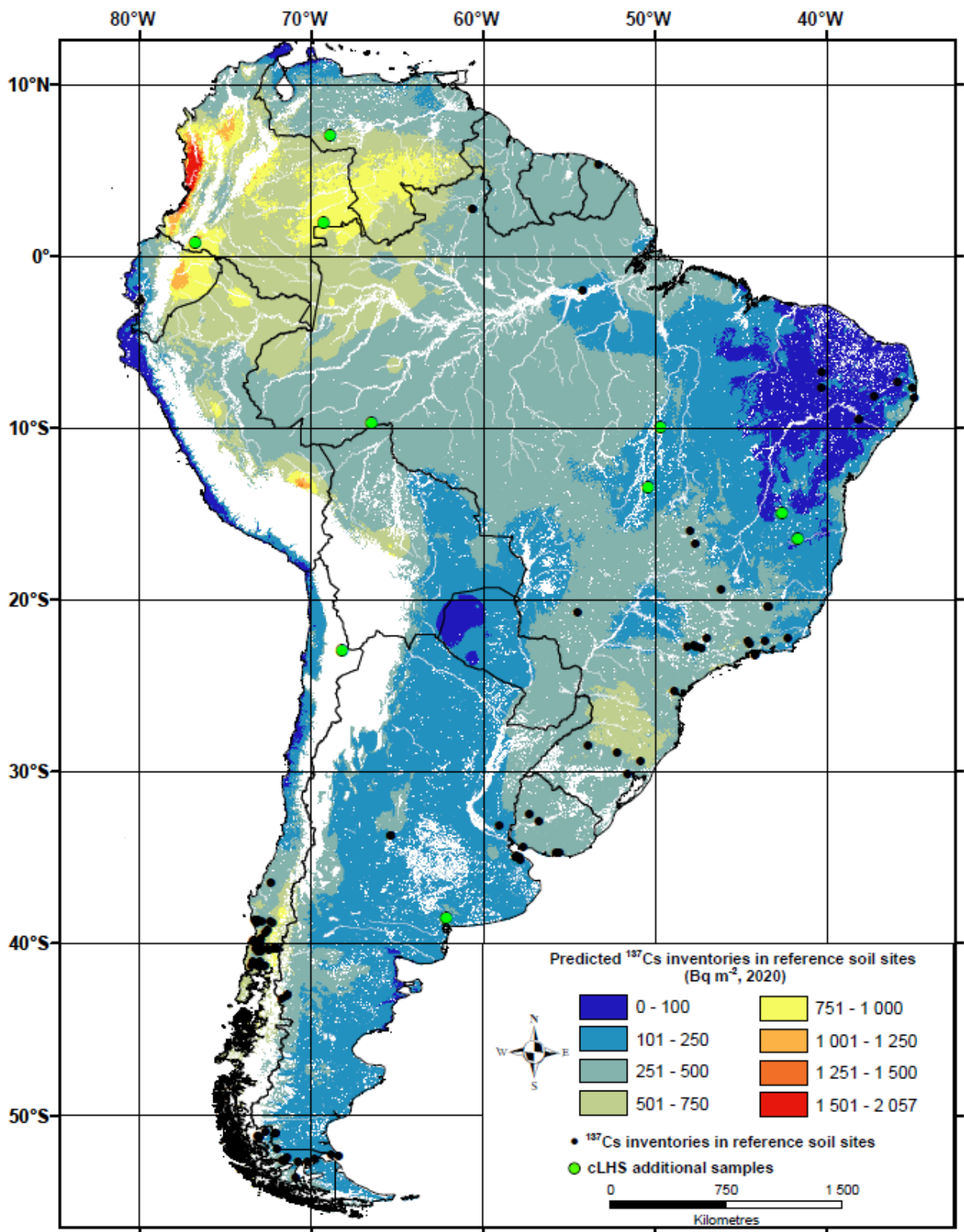


Figure 7: Baseline ^{137}Cs inventories in reference soil sites (Bq m^{-2} , 2020) estimated by Partial Least Square Regression (PLSR) with a spatial resolution of 2 km. Black dots represent those ^{137}Cs inventories compiled in the literature and measured by the authors, while green dots represent 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).

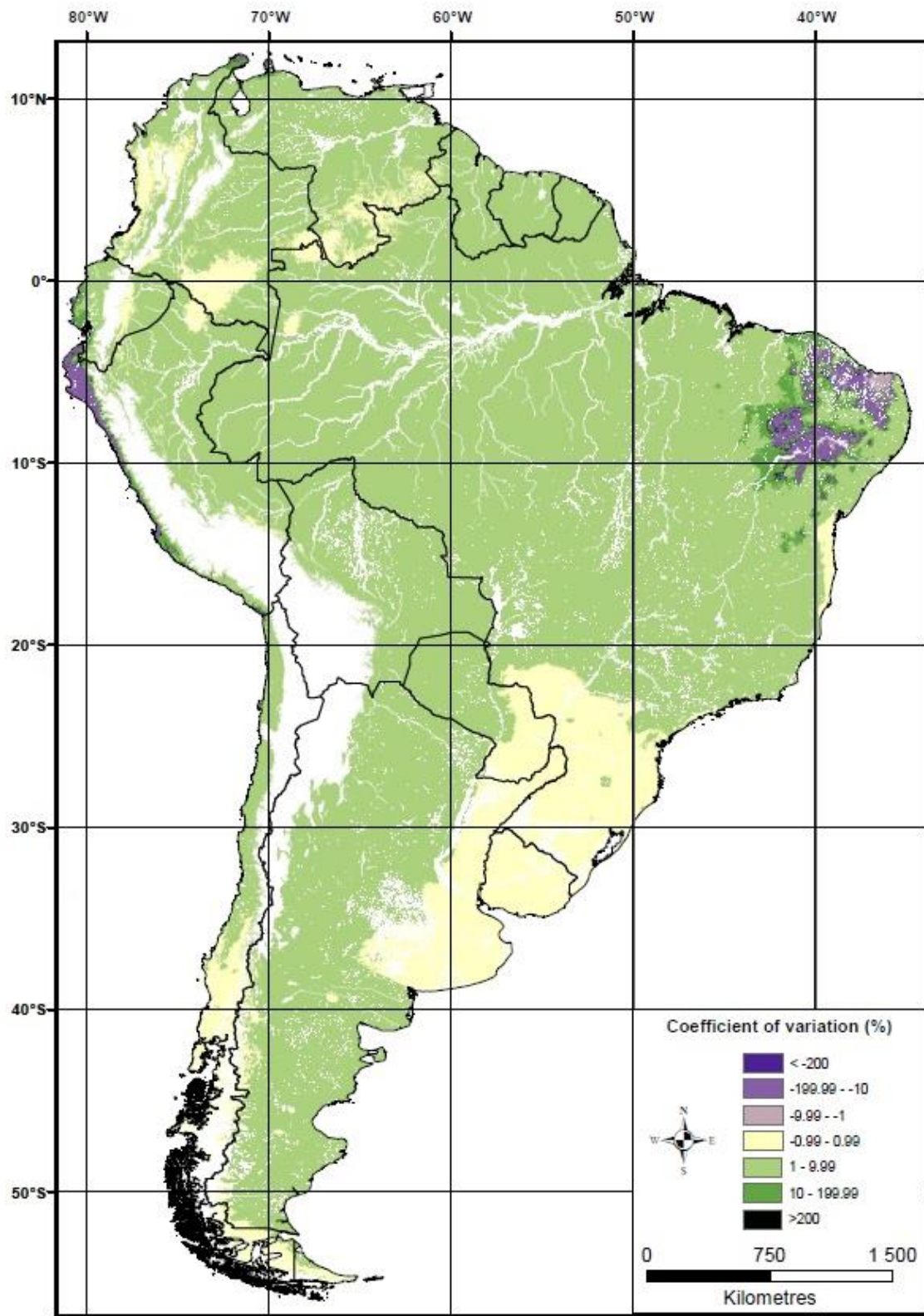


Figure 8: Prediction uncertainties of the baseline map of ^{137}Cs inventories in reference soil sites assessed through a non-parametric bootstrap approach ($n= 100$ contributing predictions). Areas above 1,800 m a.s.l., glaciers and large water bodies have been masked in white using the GMTED2010 30-arc-second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

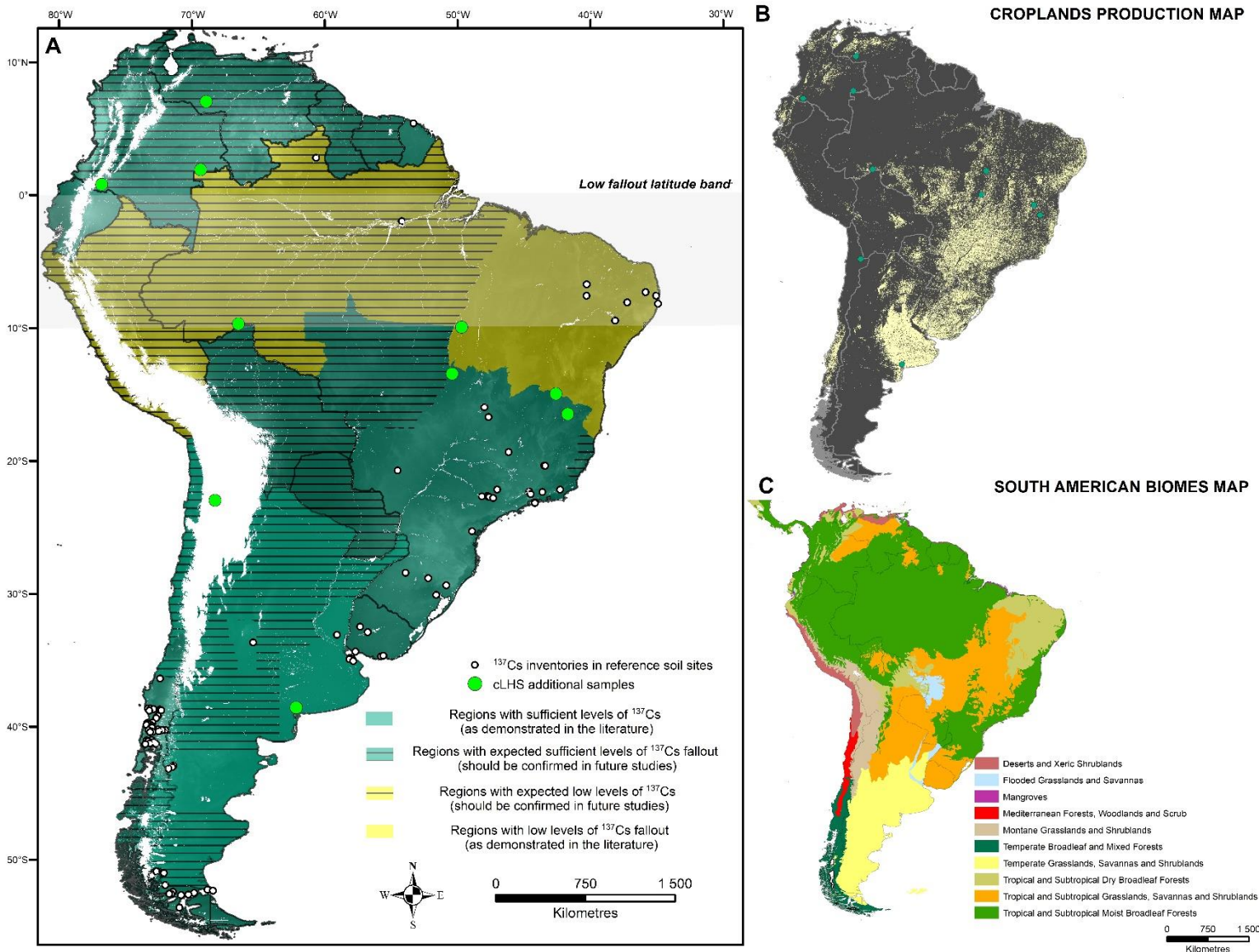


Figure 9: (A) Potential use of ^{137}Cs fallout inventories for Earth Science applications in South America. Areas above 1,800 m a.s.l., glaciers and large water bodies have been masked in white using the GMTED2010 30-arc-second elevation database (Danielson and Gesch, 2011) and the GLIMS glacier database (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)