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## Mapping the spatial distribution of global $^{137}\text{Cs}$ fallout in soils of South America as a baseline for Earth Science studies

Pierre-Alexis Chaboche, Nicolas Saby, J. Patrick Laceby, Jean P.G. Minella, Tales Tiecher, Rafael Ramon, Marcos Tassano, Pablo Cabral, Mirel Cabrera, Yuri Jacques Agra Bezerra da Silva, et al.

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

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24

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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10 29 derived fallout radiocesium ( $^{137}\text{Cs}$ ) bound to fine particles is one of the few techniques available to  
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12 30 reconstruct soil redistribution rates and evaluate the sustainability of farming practices over the recent  
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14 31 phase of agricultural intensification (1960s–2020). However, information about the spatial distribution  
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16 32 of  $^{137}\text{Cs}$  fallout across the soils of South America remains scarce, and the published data has not been  
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18 33 synthesized at the scale of this subcontinent so far. The objective of the current research is therefore  
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21 34 to quantify and map the initial  $^{137}\text{Cs}$  fallout at the scale of South America, based on the compilation of  
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23 35 published  $^{137}\text{Cs}$  inventories, additional measurements conducted on undisturbed soil profiles and  
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25 36 digital soil mapping as this baseline information may be useful for a wide range of Earth Science  
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27 37 applications. A database of  $^{137}\text{Cs}$  inventories at 96 reference sites (i.e. areas without soil erosion nor  
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30 38 accumulation) has been compiled for a variety of soil profiles (Argentina = 10, Brazil = 34, Chile = 46,  
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32 39 Uruguay = 5, French Guiana = 1) located between 5.3° North latitude and 53° South latitude. The spatial  
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34 40 distribution of  $^{137}\text{Cs}$  fallout was shown to be highly latitude-dependent, with a maximum in the 30-50°  
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36 41 South latitude band. There were higher fallout levels than expected between 20 to 60° South latitude  
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39 42 compared to the previous estimations made by UNSCEAR. A partial least square regression approach  
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41 43 based on rainfall data and geographical information as covariates was used to create a baseline map  
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43 44 of  $^{137}\text{Cs}$  fallout in soils of continental South America. This baseline map provides a powerful reference  
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45 45 dataset to anticipate the order of magnitude of  $^{137}\text{Cs}$  inventories in undisturbed soil profiles collected  
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47 46 in Brazil and Southern Chile and for numerous other applications in Earth Sciences. The potential  
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50 47 application of the  $^{137}\text{Cs}$  inventory technique in countries of South America in general, and in regions  
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52 48 with vulnerable ecosystems threatened by the expansion of agricultural activities in particular, is  
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54 49 discussed in light of this comprehensive literature review. Furthermore, the regions (i.e. the North-  
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56 50 Western part of the continent) where additional samples should be collected in priority to improve  
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59 51 this baseline map are outlined. Our results demonstrate that  $^{137}\text{Cs}$  inventories are sufficiently high to  
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52 investigate soil redistribution rates in most of South American countries where detectable levels of  
53  $^{137}\text{Cs}$  can be expected to be found in sites exposed to erosion.

54 **Keywords:** Digital soil mapping, Caesium-137, Soil erosion, Soil redistribution rates, Reference soil sites  
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## 56 **1. Introduction**

57

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

70 In order to evaluate the sustainability of agricultural practices and promote effective soil management  
71 practices, a fundamental prerequisite is to quantify soil erosion rates. Measuring soil inventories of  
72 fallout radionuclide Caesium-137 ( $^{137}\text{Cs}$ ,  $t_{1/2} = 30.2$  years) has shown great potential to provide  
73 retrospective information on soil redistribution rates over the medium-term (Ritchie and Ritchie,  
74 2007), despite the critiques made on this method (Parsons and Foster, 2011). The latter authors  
75 recommended that future studies should present their results in light of the underlying hypotheses  
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 <sup>137</sup>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 <sup>137</sup>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, <sup>137</sup>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 <sup>137</sup>Cs inventories (Bq m<sup>-2</sup>) 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 <sup>137</sup>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 <sup>137</sup>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  
2 103  
3 distribution of initial  $^{137}\text{Cs}$  fallout worldwide (Owens and Walling, 1996; Ritchie and McHenry, 1990;  
4 104  
5 Walling and He, 2000). The deposition of fallout radionuclides onto the Earth surface is conditioned by  
6 105  
7 two major factors: latitude and rainfall. Ground deposition of radionuclides from the global fallout was  
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9 shown to be highly latitude-dependent, with a maximum in the 40-50° bands in both hemispheres  
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11 (UNSCEAR, 2000). In addition, the activity of  $^{137}\text{Cs}$  deposited on the ground and its variation from one  
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13 location to another is closely linked to the annual rainfall rates, as a consequence of precipitation  
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15 scavenging (Bouisset et al., 2018; La Manna et al., 2019; Le Roux et al., 2010; Malakhov and Pudovkina,  
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17 1970). Moreover, relationships between monthly rainfall rates and  $^{90}\text{Sr}$  fallout were observed in South  
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19 America, with a peak during the months of June-August (Volchok, 1965; Volchok, 1966). Based on these  
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21 two factors, digital mapping using geostatistical approaches has been conducted at different scales to  
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23 map bomb-derived  $^{137}\text{Cs}$  fallout (Almgren et al., 2006; Aoyama et al., 2006; Chappell et al., 2011a;  
24 114  
25 Furuichi and Wasson, 2013; Meusburger et al., 2020; Palsson et al., 2006). Although reconstructions  
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27 of baseline  $^{137}\text{Cs}$  inventories were made for Europe, Australia and Eastern Asia, information about the  
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29 spatial distribution of  $^{137}\text{Cs}$  in soils of South America remains scarce, which requires further  
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31 investigation to determine the potential for application of  $^{137}\text{Cs}$  for Earth Science studies in this  
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33 subcontinent. Of note, the  $^{137}\text{Cs}$  inventories in soils of the Southern hemisphere are usually sufficient  
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35 to be measurable by gamma spectrometry when using appropriate low-background detectors during  
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37 sufficient counting times (FAO/IAEA, 2017).

48 122 Quantifying  $^{137}\text{Cs}$  inventories in soils and calculating the corresponding soil redistribution rates during  
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50 the intensive cultivation period (1960 to present) is particularly important to evaluate the sustainability  
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52 of agricultural practices implemented in South America during the last several decades to maintain  
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54 erosion at sustainable levels (Minella et al., 2014). During the last 50 years, most countries of the  
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56 subcontinent have abandoned conventional tillage to implement no-till farming (Montgomery, 2007).  
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58 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  $^{137}\text{Cs}$  inventories in reference soil sites of  
136 South America is therefore required to assess soil redistribution rates with the  $^{137}\text{Cs}$  technique. Based  
137 on the compilation of published  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  inventories in reference soil sites across South America, (2) to map the initial  $^{137}\text{Cs}$   
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  $^{137}\text{Cs}$  global fallout.

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

### 2.1. Global fallout following nuclear weapon tests

152 The UNSCEAR reports provide the main source of information regarding  $^{137}\text{Cs}$  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 <sup>137</sup>Cs reached a peak in 1963 and 1965  
2 156 tests of atmospheric nuclear devices in 1962, the deposition of <sup>137</sup>Cs reached a peak in 1963 and 1965  
3  
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  
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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  $^{90}\text{Sr}$  fallout  
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14 186 sampling, was the largest and the most widely distributed and therefore, it has been adopted by the  
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16 187 UNSCEAR to estimate the total hemispheric annual deposition of  $^{90}\text{Sr}$  (UNSCEAR, 1993). The global  
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18 188 distribution of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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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29 192 of data is lacking for monitoring stations ( $n=34$ ) located in South America. In all, 59% of monthly  $^{90}\text{Sr}$   
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31 193 fallout data is missing between 1954-1976, as most of the monitoring stations did not have continuous  
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33 194 records (Health and Safety Laboratory, 1977). In addition, when studies investigating  $^{137}\text{Cs}$  inventories  
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36 195 in undisturbed soil profiles are conducted at regional or continental scales, the proportion of global  
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38 196 fallout estimated by UNSCEAR appears to be underestimated (Aoyama et al., 2006; Chappell et al.,  
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41 197 2011a; Schuller et al., 2002; Tagami et al., 2019). Regarding the information currently available for  $^{137}\text{Cs}$   
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43 198 fallout at the scale of South America, the only reference is a study published by Garcia Agudo (1998)  
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45 199 who also used results of these monitoring programmes to map the global inventory of  $^{137}\text{Cs}$  from  
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48 200 nuclear tests worldwide. Given the wide range of applications of  $^{137}\text{Cs}$  in the scientific literature, as  
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50 201 well as the debate raised on its use as a tracer for soil erosion assessment (Mabit et al., 2013; Parsons  
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52 202 and Foster, 2011; Parsons and Foster, 2013), a more accurate characterization of the initial fallout of  
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55 203 bomb-derived  $^{137}\text{Cs}$  and its spatial distribution across South America is fundamental to provide a  
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57 204 baseline for future studies.

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206 2.2. Data collection on <sup>137</sup>Cs in reference soil sites of South America

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

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

221  
222 where  $\lambda$  is the decay constant of <sup>137</sup>Cs ( $\lambda = \ln 2 / 30.2 \text{ y}$ ) and  $t$  is the time (year) since the sampling year  
223 as reported in the corresponding article or estimated following the above-mentioned method. Each  
224 entry in the database corresponds to one undisturbed soil profile for which the <sup>137</sup>Cs inventory has  
225 been calculated, its geographical coordinates (latitude, longitude, elevation), annual rainfall rates,  
226 sampling method, <sup>137</sup>Cs inventory and the associated standard deviation estimated by the author and  
227 the associated decay-corrected values calculated from Eq. (1). In total, a database of 103 <sup>137</sup>Cs  
228 inventories at reference soil sites has been compiled, in four different countries and one overseas

229 department of France (Antarctica = 2, Argentina = 10, Brazil = 38, Chile = 50, Uruguay = 2, French Guiana  
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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 <sup>137</sup>Cs inventories calculated at these reference soil sites, respectively.  
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9 233 Four <sup>137</sup>Cs 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<sup>-2</sup> respectively) and in Southern Brazil (5 and 90 Bq m<sup>-2</sup>), 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 <sup>137</sup>Cs inventories were found  
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19 238 in the central part of Chile (3113 and 2860 Bq m<sup>-2</sup>), at 40° South latitude (Schuller et al., 2002). These  
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21 239 inventories, with elevated <sup>137</sup>Cs 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 <sup>137</sup>Cs fallout derived from this dataset,  
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31 242 additional unpublished <sup>137</sup>Cs 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 <sup>137</sup>Cs (662 keV)  
40  
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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48 250 among the 103 <sup>137</sup>Cs inventories compiled in the literature and the additional three <sup>137</sup>Cs inventories  
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50 251 collected by the co-authors, 96 <sup>137</sup>Cs inventories (Table 1) were used to perform the statistical analysis  
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53 252 presented below.  
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254 2.3. Spatial distribution of <sup>137</sup>Cs inventories in South America

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256 According to the current literature review, the highest <sup>137</sup>Cs 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 <sup>137</sup>Cs inventories were 97 and 279  
259 Bq m<sup>-2</sup>, respectively. The distribution of <sup>137</sup>Cs 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 <sup>137</sup>Cs 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 <sup>137</sup>Cs inventories in  
265 undisturbed soil profiles is mostly valid for Brazil, Uruguay, Argentina and Chile.

266 The typical pattern of <sup>137</sup>Cs 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 <sup>137</sup>Cs  
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, <sup>137</sup>Cs 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  $^{137}\text{Cs}$  inventories in reference soil sites and annual  
1  
2 279 rainfall rates provided by the authors ( $R^2=0.43$ ,  $n=92$ ,  $p < 0.001$ ). However, a wide dispersion of  $^{137}\text{Cs}$   
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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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6  
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  $^{137}\text{Cs}$  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 ( $R^2=0.39$ ,  $n=38$ ,  $p < 0.001$ ).  
14  
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16  
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  $^{137}\text{Cs}$  inventories ( $n=74$ ). However,  
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21  
22 287 when the analysis is performed for  $^{137}\text{Cs}$  inventories in reference soil sites located at the same latitude  
23  
24 288 although at a different altitude, it can be observed that  $^{137}\text{Cs}$  inventories tends to increase with the  
25  
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  $23^\circ$  and  $29^\circ$  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  $^{137}\text{Cs}$   
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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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40  
41 295  $p < 0.01$ ). A step wise multiple regression indicated a significant relationship between observed and  
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43 296 predicted  $^{137}\text{Cs}$  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 $^{137}\text{Cs}$ 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 <sup>137</sup>Cs  
306 fallout (Table 2). The spatial distribution of <sup>137</sup>Cs 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 <sup>137</sup>Cs deposition data from the Arctic Monitoring and  
309 Assessment Programme (AMAP) with annual rainfall rates for the period 1955-1985 to predict <sup>137</sup>Cs  
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 <sup>137</sup>Cs measurements in undisturbed soil  
312 profiles, to predict the spatial variation in global fallout of <sup>137</sup>Cs 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 <sup>137</sup>Cs 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.

319

### 320 3.2. Digital mapping of <sup>137</sup>Cs 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 <sup>137</sup>Cs 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, <sup>137</sup>Cs 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  
1  
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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9  
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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24  
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 <sup>137</sup>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 <sup>137</sup>Cs variability observed and tended to underestimate inventories  $> 800 \text{ Bq m}^{-2}$ . The predicted  
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52 352 <sup>137</sup>Cs inventories followed a right-skewed statistical distribution, with an average of  $348 \text{ Bq m}^{-2}$  and a  
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55 353 standard deviation of  $237 \text{ Bq m}^{-2}$ . Highest values (between  $1501$  to  $2057 \text{ Bq m}^{-2}$ ) were found in  
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57 354 Colombia and few areas of the Andean Cordillera, while the vast majority of <sup>137</sup>Cs inventories lower  
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59 355 than  $100 \text{ Bq m}^{-2}$  were found in the North-Eastern part of Brazil (Fig. 7). Surprisingly, high <sup>137</sup>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 <sup>137</sup>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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16 363 2011). As previously mentioned, most of the <sup>137</sup>Cs inventories in undisturbed soil profiles used to  
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18 364 develop the digital soil mapping approach were measured in the eastern part of South America and in  
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21 365 Southern Chile. The estimates provided for the other regions (i.e. northern and western parts of the  
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23 366 subcontinent) should therefore be interpreted with caution. Additional sampling in these areas will be  
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26 367 necessary to improve model predictions of <sup>137</sup>Cs global fallout in South America.

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29 368 The baseline map of <sup>137</sup>Cs inventories in reference soil sites (Bq m<sup>-2</sup>, 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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36 371 levels of fallout <sup>137</sup>Cs at the subcontinental-scale of South America relies on the assumption that studies  
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38 372 listed in our database followed the recommendations to select and sample undisturbed soil sites. Since  
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41 373 the coefficient of variation of multiple samples collected to characterize one reference site is not  
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43 374 always reported by authors, the observations uncertainties are difficult to quantify. As reported in the  
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45 375 literature, <sup>137</sup>Cs inventories in reference soil sites have a coefficient of variation of approximately 20%,  
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48 376 as a consequence of random and systematic spatial variability, sampling variability and measurement  
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50 377 precisions (Loughran et al., 2002; Owens and Walling, 1996; Pennock, 2000). Accordingly, the map  
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52 378 predictions provided in this study for each grid should be interpreted as a trend rather than a single  
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54  
55 379 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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6  
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  $^{137}\text{Cs}$  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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15 388 coefficient of variation (in %) was computed by dividing the average prediction by the 95% confidence  
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17 389 interval (Fig. 8). For most part of South America, the relative error was comprised between 0 and 10  
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19 390 %. The highest uncertainties could be observed in two areas located in the North-Eastern part of Brazil  
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21 391 and along the Pacific coast of Peru, where the map predictions of  $^{137}\text{Cs}$  inventories in reference soil  
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23 392 sites are the lowest (0 to 100 Bq m<sup>-2</sup> 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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41 399 expected to provide a scheme of representative samples covering the study area with a relatively small  
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43 400 sample size for financial and logistical reasons. If it is assumed that the soil property is linked to  
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45 401 environmental covariates, a robust strategy is to ensure that the measurements are also uniformly  
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47 402 spread in the feature (i.e. covariates) space. However, the collected data of the present study did not  
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49 403 follow any sampling design as they were compiled from various independent studies, leading to large  
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51 404 areas devoid of sampling observations or places with clusters. Accordingly, an experiment was  
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53 405 conducted to generate new sampling locations in order to fill the gap in feature space of the selected  
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55 406 environmental variables. This can be achieved using conditioned Latin Hypercube sampling (cLHS)  
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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 $^{137}\text{Cs}$ technique in South America

416 In South America, where the input of bomb-derived  $^{137}\text{Cs}$  fallout has stopped since the 1980s,  $^{137}\text{Cs}$   
417 inventories in soils are continuously decreasing as a consequence of radioactive decay. One major  
418 challenge is to clearly identify areas where the  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  which would  
421 therefore prevent the application of the  $^{137}\text{Cs}$  technique in these locations.

422 Chile is the country of South America where information about  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  inventory of 563 Bq m<sup>-2</sup>  
425 can be calculated from 30 reference soil sites. The high content of  $^{137}\text{Cs}$  in soils allowed for the use of  
426 the  $^{137}\text{Cs}$  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<sup>-2</sup> 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<sup>-1</sup> without any information regarding the bulk density of soils sampled for calculating  
430 the other  $^{137}\text{Cs}$  inventories collected in the country (Schuller et al., 2002). Although  $^{137}\text{Cs}$  inventories  
431 are lower in the Patagonian part of Chile with a mean value of 280 Bq m<sup>-2</sup> ( $n=16$ ), the applicability of  
432 the  $^{137}\text{Cs}$  technique could be explored to investigate the soil response to environmental changes under  
433 colder climates (Navas et al., 2018).

434 In Argentina, <sup>137</sup>Cs soil profiles have been collected in the Pampa Ondulada region of Buenos Aires  
1  
2 435 Province (Bujan et al., 2003), in soils of La Plata region (Montes et al., 2013), in natural and semi-natural  
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4 436 grassland areas of San Luis Province (Ayub et al., 2008) and in the Patagonian Andean forests (La  
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7 437 Manna et al., 2019). These studies showed that <sup>137</sup>Cs inventories were sufficient in these regions to  
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9 438 calculate soil redistribution rates based on this technique. Of note, there is a lack of <sup>137</sup>Cs inventory  
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11 439 data in the literature for the Argentinian provinces located between 25 to 40° South latitudes, where  
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13 440 inventories in reference soil sites should be the highest of South America according to UNSCEAR  
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15 441 (2000). Thus, the <sup>137</sup>Cs technique should be successfully implemented in this country exposed to  
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17 442 elevated levels of both water and wind erosion (Ares et al., 2016; Mendez and Buschiazzo, 2010).  
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22 443 In Uruguay, only two <sup>137</sup>Cs inventory had been documented so far in the literature (Alonso et al., 2012;  
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24 444 Tassano et al., 2020). Alonso et al. (2012) assessed soil erosion rates in a forested micro-catchment  
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26 445 occupied by eucalyptus plantations within the Río Negro River basin. The three additional <sup>137</sup>Cs  
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28 446 inventories collected in Uruguay by co-authors of the current research remained in the same order of  
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30 447 magnitude, with an average value of  $336 \pm 13 \text{ Bq m}^{-2}$ . These values are consistent with <sup>137</sup>Cs inventories  
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32 448 found in the neighbouring Rio Grande do Sul state, in southernmost Brazil, where <sup>137</sup>Cs inventories  
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34 449 followed an increasing gradient from the Uruguayan border in the South ( $315 \pm 22 \text{ Bq m}^{-2}$ ) to the North  
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36 450 ( $1022 \pm 292 \text{ Bq m}^{-2}$ ) (Didone et al., 2019; Handl et al., 2008; Minella et al., 2014).  
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41 451 In general, <sup>137</sup>Cs inventories measured in sites of Brazil located below 20°S are high enough to  
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43 452 reconstruct soil redistribution rates (Bacchi et al., 2000; Correchel et al., 2005; Didone et al., 2019;  
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45 453 Macêdo, 2009; Minella et al., 2014). In addition to these studies where the <sup>137</sup>Cs technique has been  
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47 454 used successfully, many regions in South America should have received enough fallout to reconstruct  
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49 455 soil redistribution rates (Fig 9.A). This includes most of the agricultural regions located in Paraguay and  
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51 456 Argentina between 20 to 45°S, as well as Mato Grosso state (Brazil) where intensification in agriculture-  
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53 457 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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$   
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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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$   
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19 466 technique was not retained in the current review because geographical information of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  activity  
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25 469 patterns with depth and land use history. A mean  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  initial fallout in equatorial regions of South America.  
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31 472 Indeed, anomalous high values of  $^{137}\text{Cs}$  activities were also observed in surface soils of Venezuela  
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33 473 (LaBrecque et al., 2001) and one  $^{137}\text{Cs}$  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 ( $1022 \pm 293$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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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11 489 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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18 492 radioactive decay,  $^{137}\text{Cs}$  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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25 495 studies using fallout radionuclides, especially in the Southern hemisphere that received only 23% of  
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27  
28 496 the total bomb-fallout emitted worldwide according to UNSCEAR (2000). In the last several decades,  
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30 497 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  $^{239}\text{Pu}$  was  
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38 500 carried out in Australia (Lal et al., 2020). In contrast to  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  inventories in reference soil sites is observed in  
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55 507 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

509 sufficient levels of  $^{137}\text{Cs}$  fallout to investigate soil redistribution rates induced by deforestation and  
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2 510 land uses changes. A better characterization of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  at the subcontinental-scale of South America. The current research demonstrates that  
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35 525 the  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  fallout is scarce. Our results indicate that  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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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2 536 Latin Hypercube sampling (cLHS), the South American continent could be subdivided into regions  
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4 537 corresponding to different biomes in which the fate of the global <sup>137</sup>Cs fallout is expected to be  
5  
6 538 homogeneous. Consequently, future sampling campaigns should strive to ensure that all biomes are  
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8 well covered with a sufficient number of soil profiles.

9 539 The map generated can be used both to validate <sup>137</sup>Cs inventories collected in the field or as a decision-  
10  
11 540 support tool to guide the implementation of the <sup>137</sup>Cs 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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15 542 applications, including the vertical transfers in soils, the circulation of air masses and ocean currents.  
16  
17 543 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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19 544 optimized through the incorporation of additional <sup>137</sup>Cs inventory measurements in South America.  
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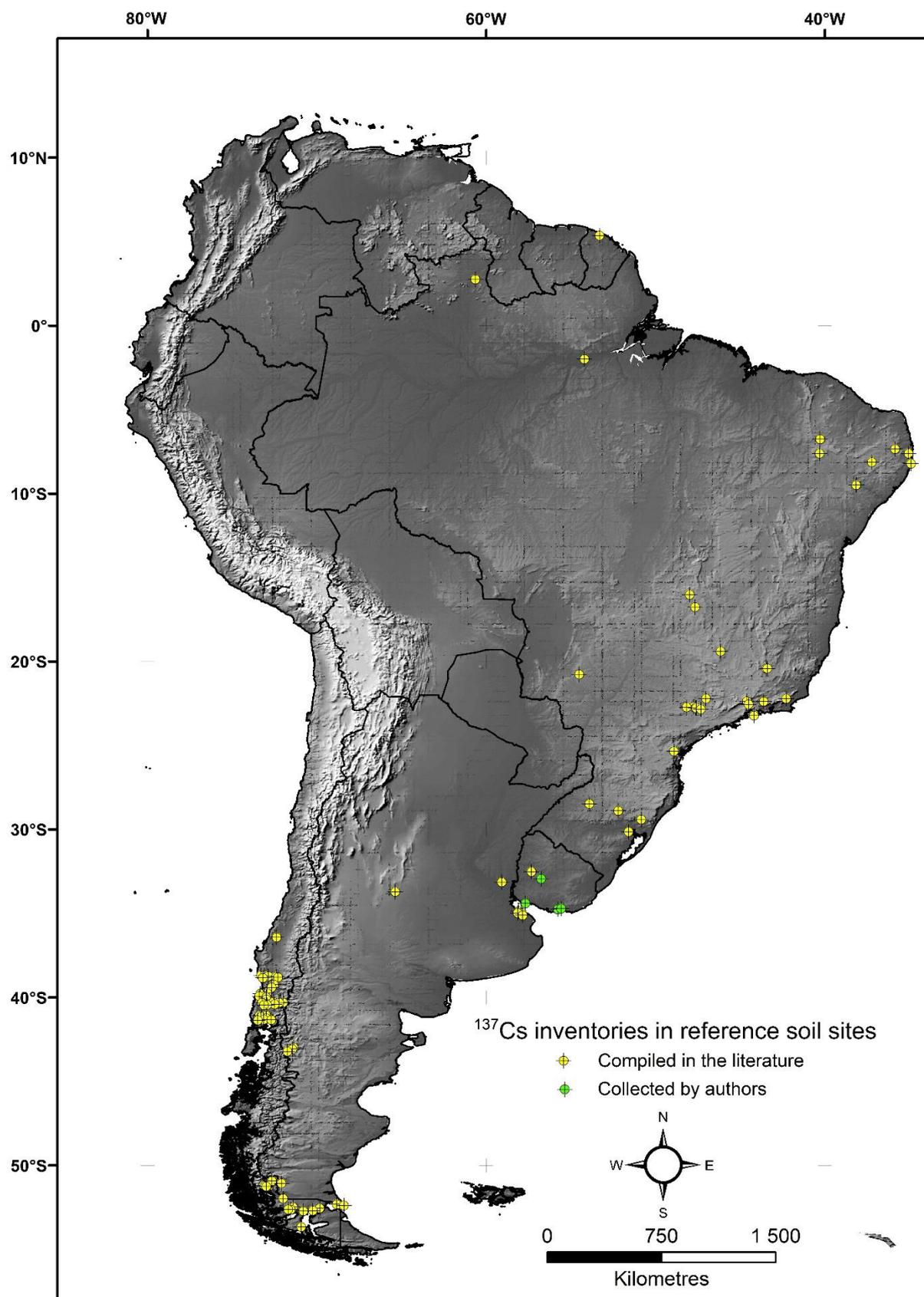
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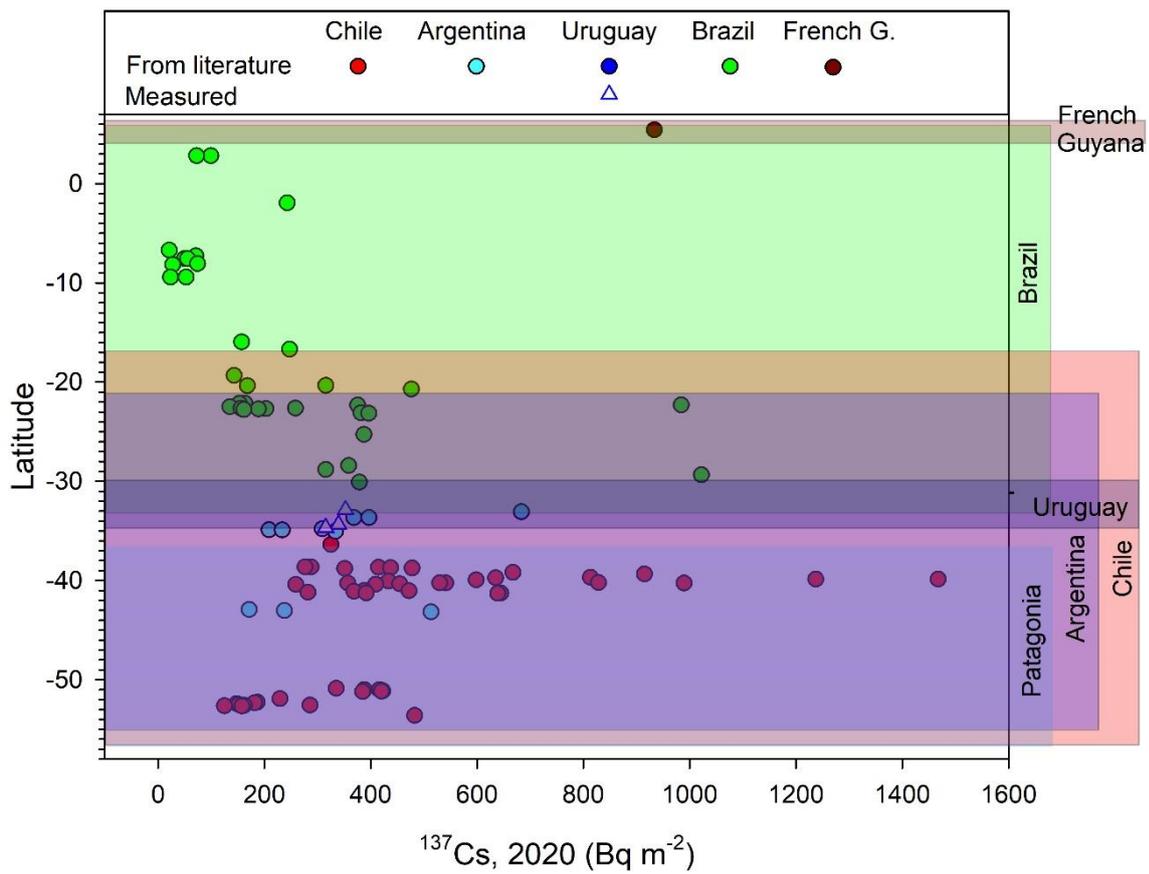
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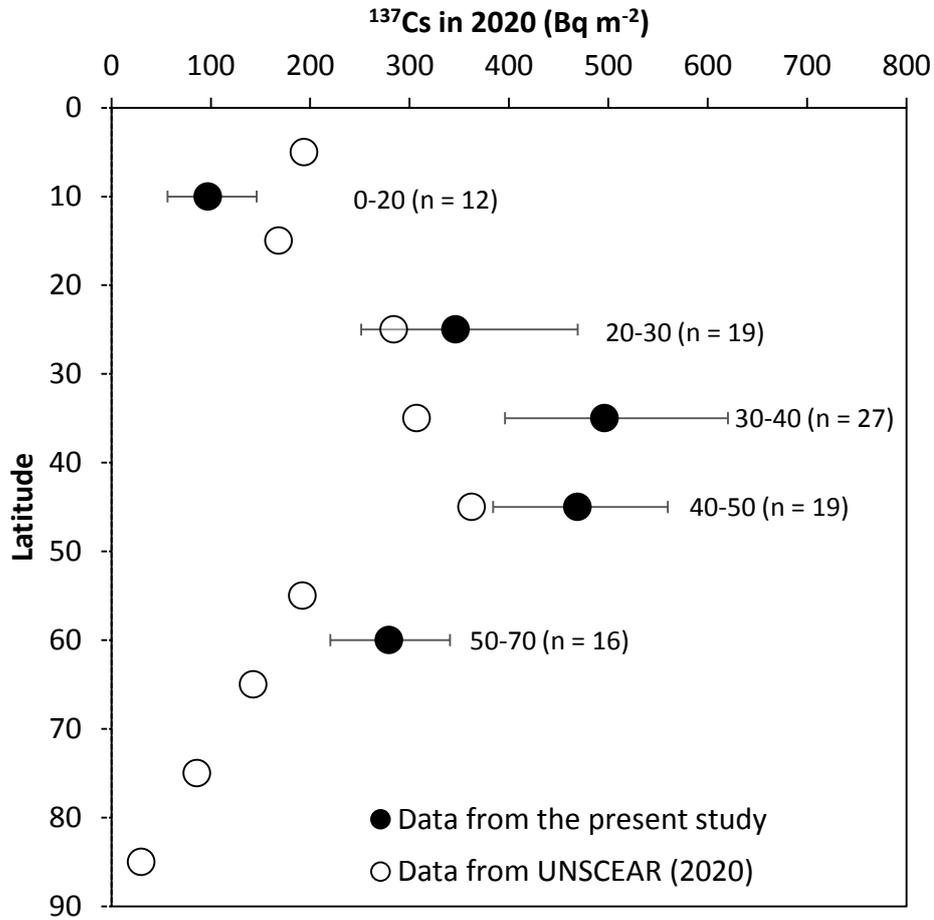
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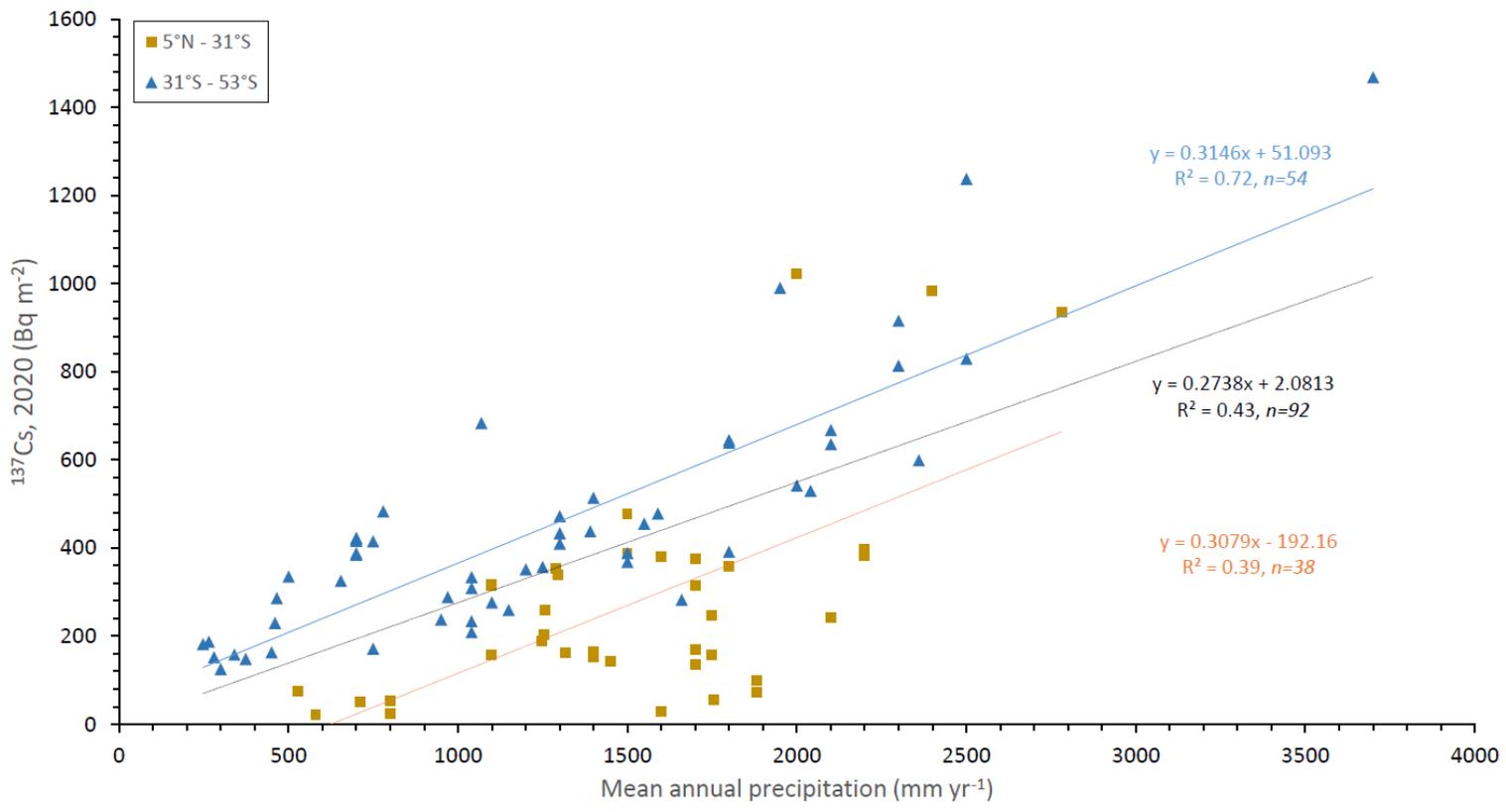
**Figure 1:** Spatial distribution of  $^{137}\text{Cs}$  reference soil sites in undisturbed soil profiles of South America as documented in the literature and collected by the authors.



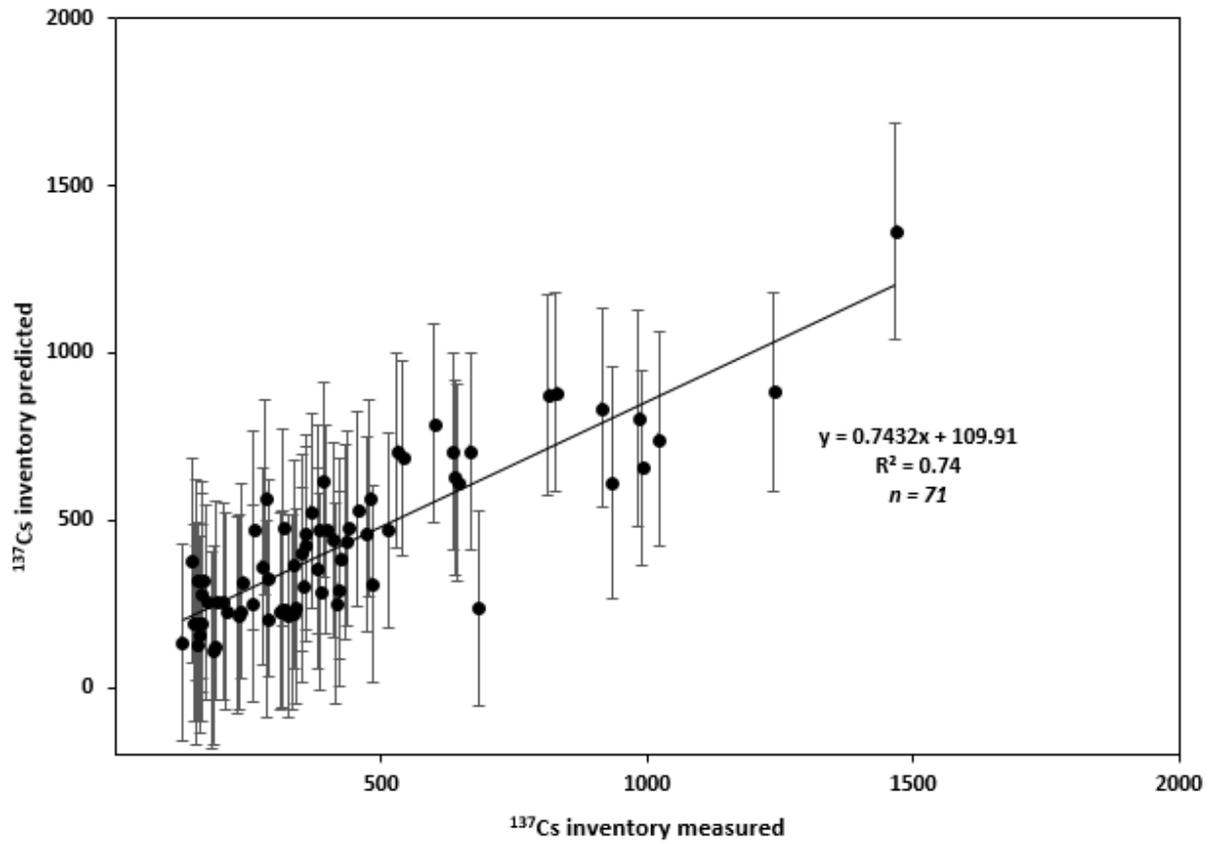
**Figure 2:** Variation of  $^{137}\text{Cs}$  inventories in reference soil sites ( $\text{Bq m}^{-2}$ ), decay-corrected to 2020 and plotted against south latitude.



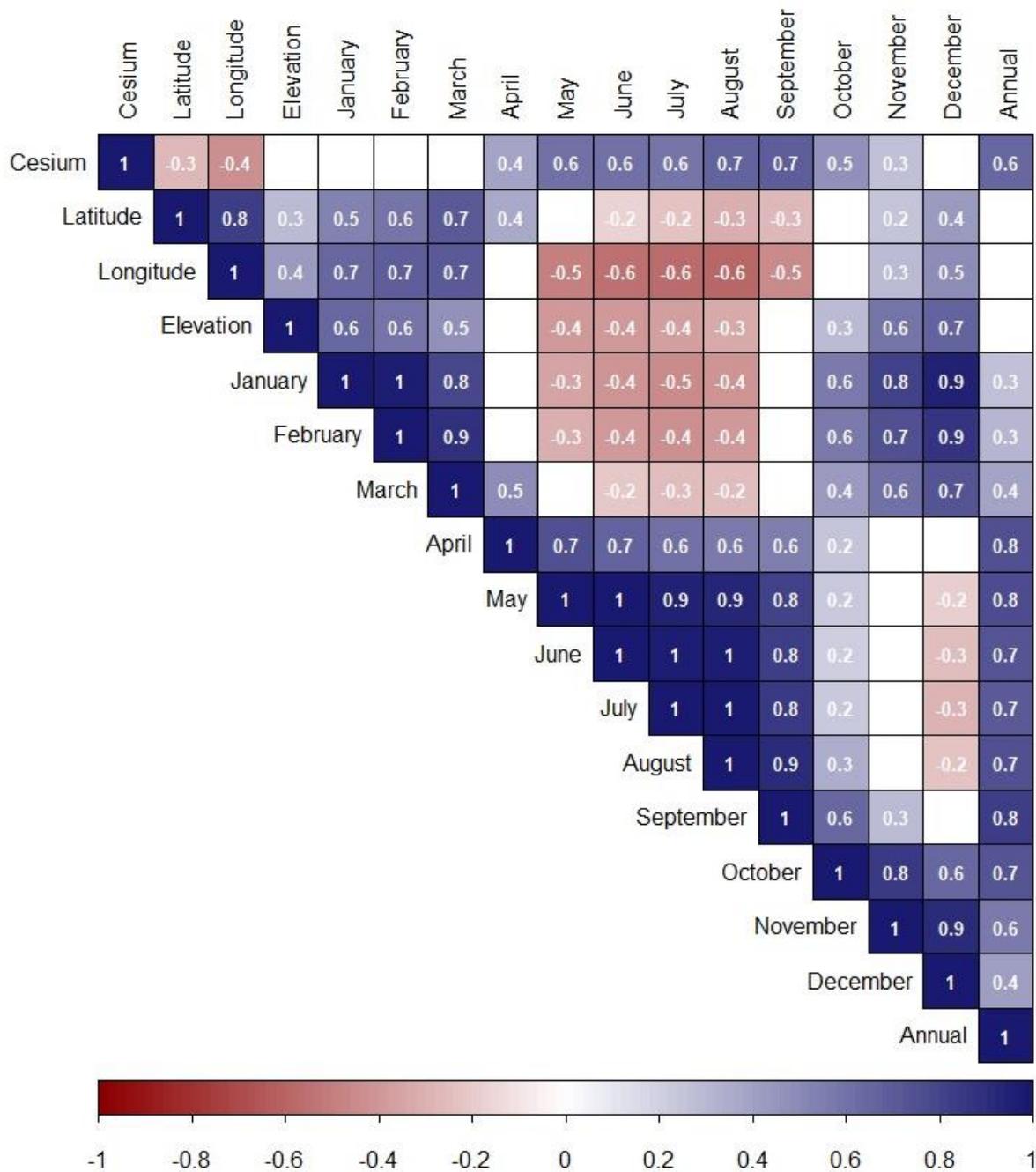
**Figure 3:** Distribution of mean  $^{137}\text{Cs}$  in reference soil sites documented in our review for each  $10^\circ$  latitude bands compared with the distribution of radionuclide fallout from thermonuclear bomb testing with latitude, after UNSCEAR (2000). This distribution was calculated with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



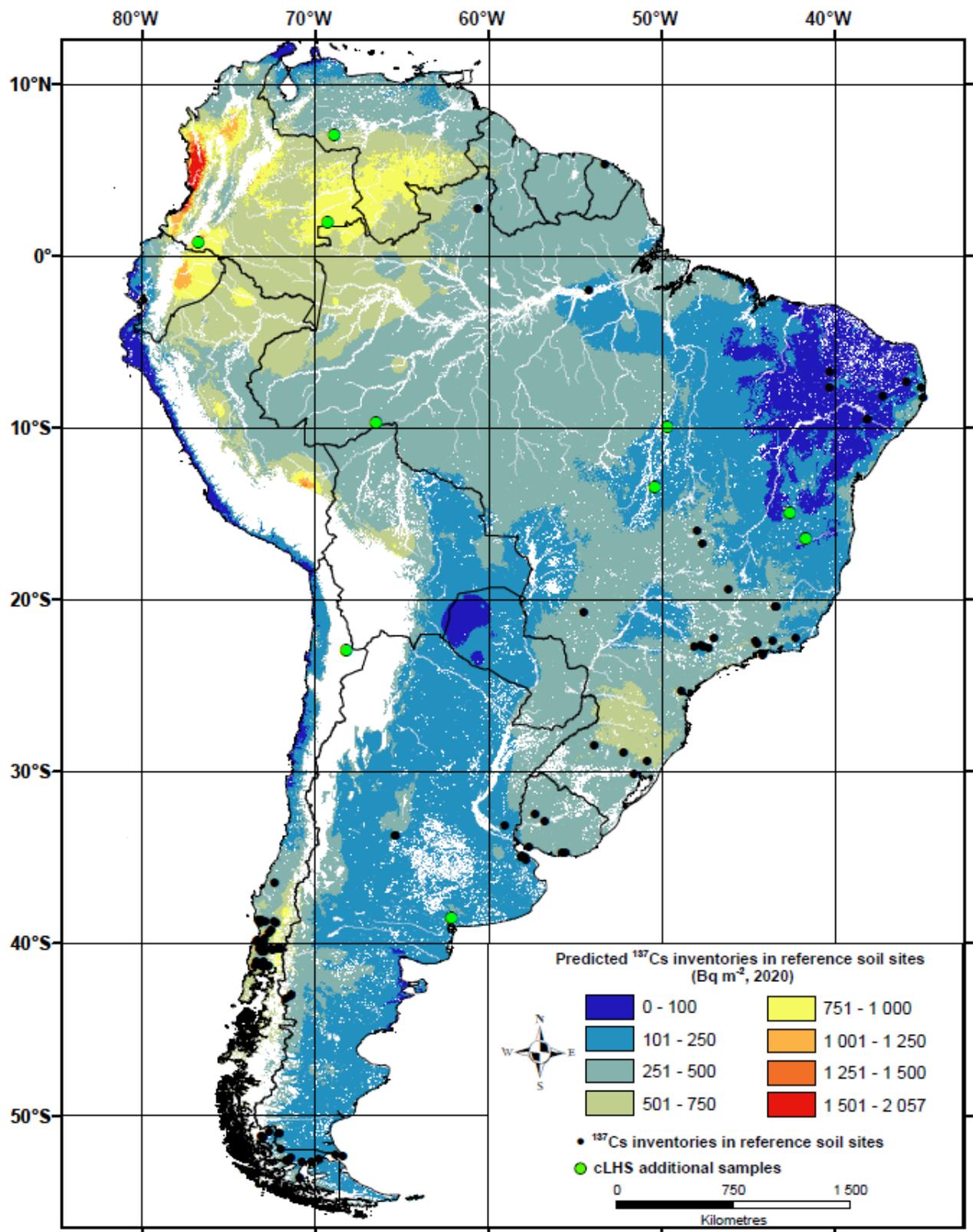
**Figure 4:** Relationships between mean annual precipitation and  $^{137}\text{Cs}$  inventories in reference soil sites for different latitude ranges.



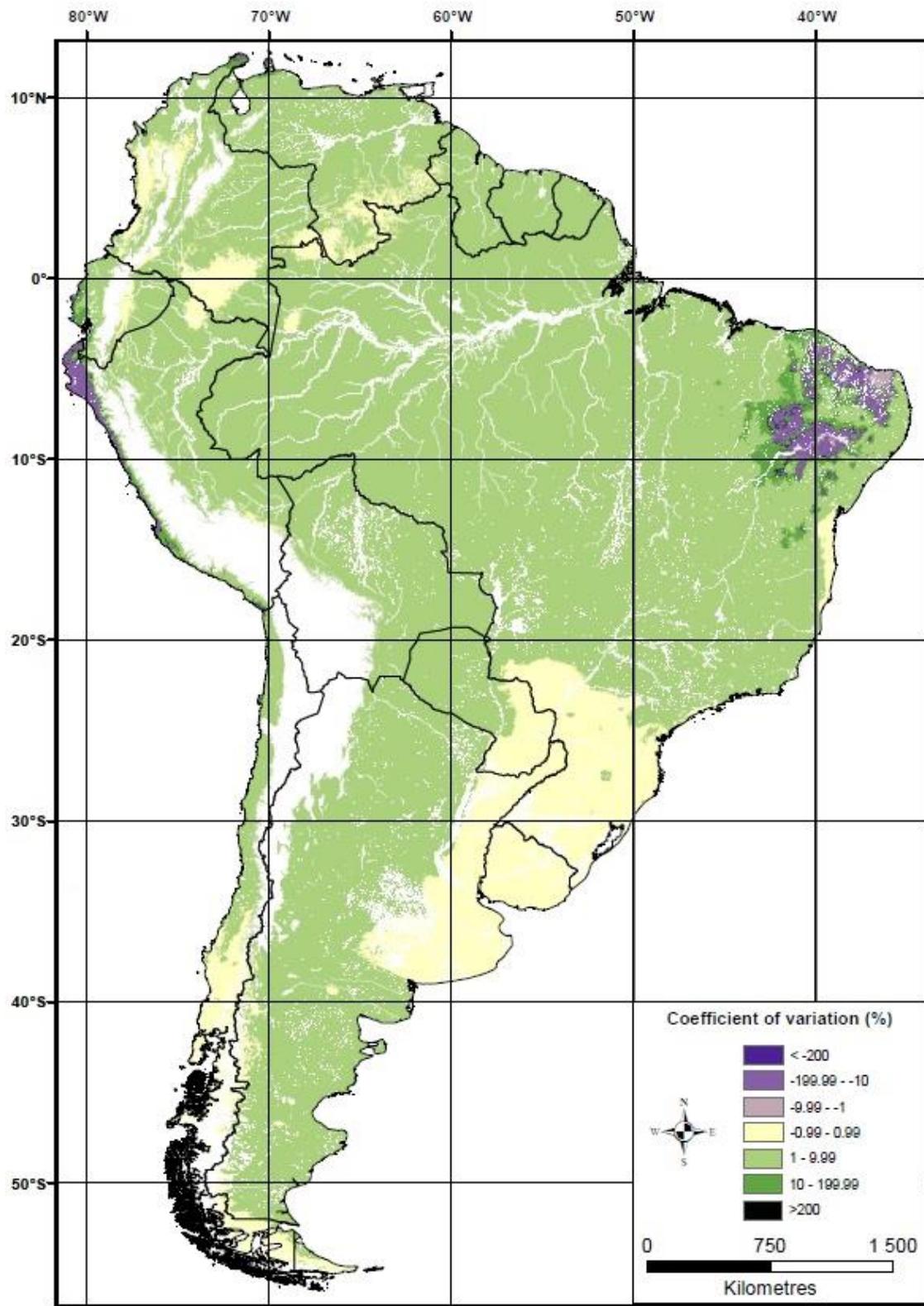
**Figure 5:** Results of the multiple regressions using latitude, longitude, elevation and rainfall plotted with the 95% confidence intervals.



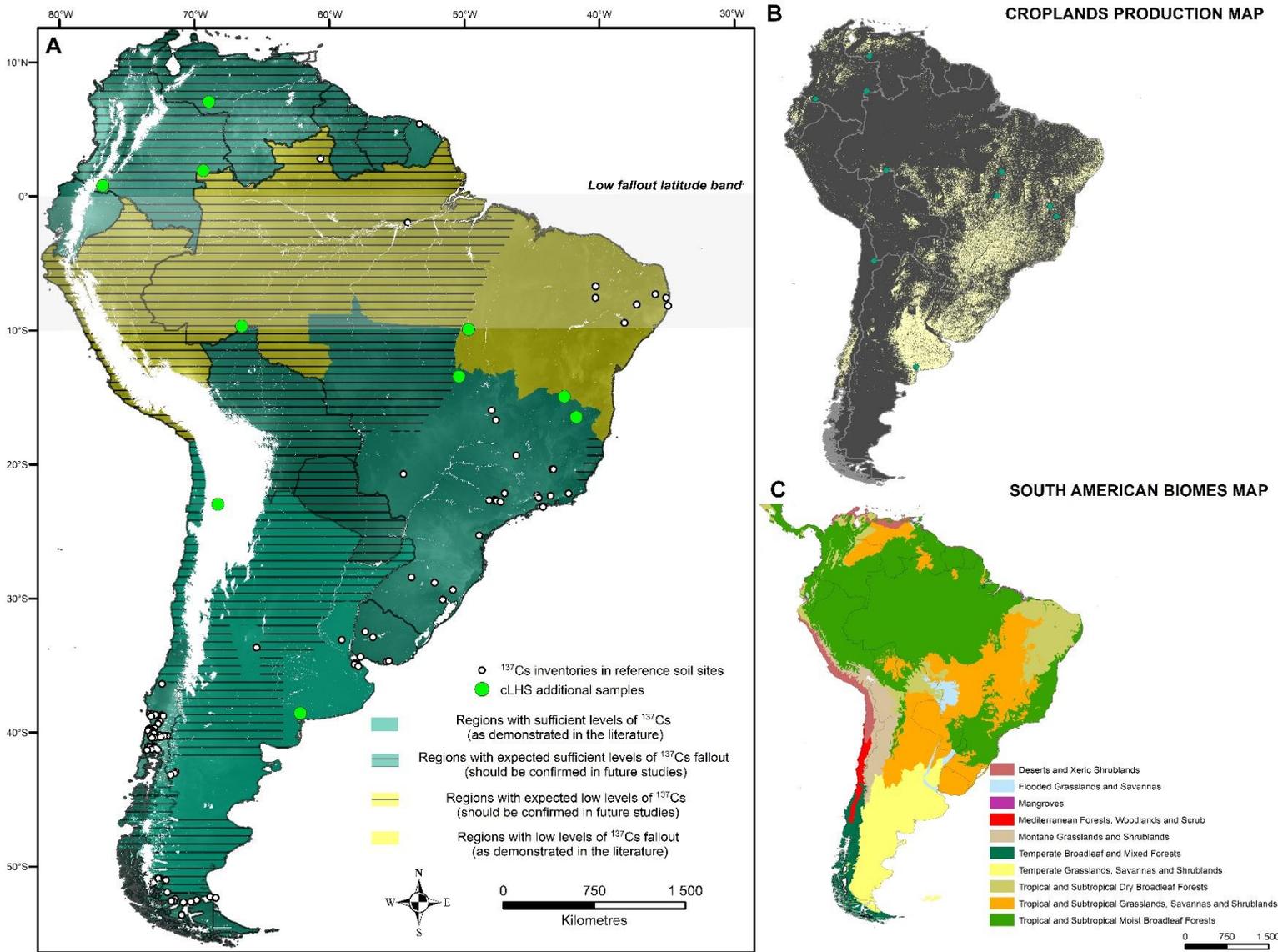
**Figure 6:** Correlogram plot for  $^{137}\text{Cs}$  inventories in reference soil sites, mean monthly/annual precipitation rates (1950-2000) and geographical information. Significant levels  $> 0.1$  are represented as white squares.



**Figure 7:** Baseline  $^{137}\text{Cs}$  inventories in reference soil sites ( $\text{Bq m}^{-2}$ , 2020) estimated by Partial Least Square Regression (PLSR) with a spatial resolution of 2 km. Black dots represent those  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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)