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## Title

$^{240}\text{Pu}/^{239}\text{Pu}$  signatures allow refining the chronology of radionuclide fallout in South America.

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## Abstract

Atmospheric nuclear tests (1945–1980) have led to radioactive fallout across the globe. French tests in Polynesia (1966–1974) may influence the signature of fallout in South America in addition to those conducted by USA and former USSR until 1963 in the Northern hemisphere. Here, we compiled the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios reported for soils of South America and conducted additional measurements to examine their latitudinal distributions across this continent. Significantly lower ratio values were found in the 20–45° latitudinal band (0.04 to 0.13) compared to the rest of the continent (up to 0.20) and attributed to the contribution of the French atmospheric tests to the ultra-trace plutonium levels found in these soils. Based on sediment cores collected in lakes of Chile and Uruguay, we show the added value of measuring  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios to refine the age models of environmental archives in this region of the world.

**Keywords:** Plutonium isotopes, Nuclear weapon tests, Geochronology, Soil erosion

## 1. Introduction

Anthropogenic radionuclides emitted by atmospheric nuclear weapon tests (NWT) conducted after 1945 have been widely used as powerful markers to date environmental archives and conduct climatic and environmental reconstructions (Alewell et al., 2017; Foucher et al., 2021; Hancock et al., 2011; Ritchie and Ritchie, 2007). This information is also used both to reconstruct soil erosion rates during the Great Acceleration (Steffen et al., 2015) – the period that started in 1950 and that is characterised by a general increase of the human imprint on global biogeochemical cycles – and for sediment source fingerprinting research (Evrard et al., 2020; Meusburger et al., 2016). Over 90% of the total energy yield (440 Mt) associated with NWT was detonated in the Northern hemisphere, during tests that were mainly carried out by the United States (US) until 1958 in the Marshall Islands and then by the Former Soviet Union (FSU) between 1961 and 1962 at the Novaya Zemlya and Semipalatinsk sites (UNSCEAR, 2008). The opening to the signature of the Partial Test-Ban Treaty on August 5, 1963 has significantly reduced global emissions of artificial radionuclides into the environment, with the end of the atmospheric tests conducted by the USA, FSU and the United Kingdom (UK). After initial tests in the Saharan Desert (1960–1966), France conducted most of its nuclear experiments at the *Centre d'Expérimentation du Pacifique* (C.E.P) in Polynesia, with 41 atmospheric tests conducted in the vicinity of Moruroa and Fangataufa atolls between 1966 and 1974. In the Northern Hemisphere, China carried out atmospheric tests in Lop Nor until 1980 (Fig. 1).

<Figure 1 here>

Depending on the type of test, its yield, firing altitude and latitude, radioactive debris attached to ambient aerosol particles were distributed between the troposphere and the stratosphere. Tropospheric debris were dispersed within the latitude band of the initial injection and followed wind-driven trajectories, before being deposited onto the soil or ocean surfaces with residence times comprised between 30-70 days (Bennett, 2002; Holloway and Hayes, 1982). Stratospheric deposition across the world accounts for the majority of global long-lived fission product residues, with atmospheric residence times comprised between 3 and 24 months depending on whether the debris were injected in the polar or the equatorial regions (UNSCEAR, 2000). In addition to the ubiquitous  $^{137}\text{Cs}$  ( $T_{1/2} = 30$  years) found across the globe, the analysis of plutonium (Pu) isotopes including  $^{239}\text{Pu}$  ( $T_{1/2} = 24\ 110$  years) and  $^{240}\text{Pu}$  ( $T_{1/2} = 6\ 561$  years) offer the opportunity to discriminate between contrasted sources of radionuclides in general, and Pu in particular. Based on the available worldwide compilations, it is currently accepted that the deposition of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  reached a peak in 1963 and 1964-1965 in the Northern and Southern hemispheres, respectively (Cambray et al., 1989; Turney et al., 2018). The delay observed between fallout peaks is attributed to the time required for allowing exchanges of nuclear fallout products between both hemispheres.

Although the fallout signatures associated with the tests conducted in the Northern Hemisphere have received a significant interest, much less is known regarding the deposition that took place in South America, e.g. in response to the nuclear tests conducted in the Southern hemisphere. Although the devices tested were less powerful than those developed by the USA and the FSU, the environmental impacts of the French nuclear experiments may have led to a distinct labelling of soils and sediments across this subcontinent. Sources of radionuclides in this part of the world are more limited, as they only include part of the emissions of the tests conducted by the i) USA and the UK in equatorial regions (Pacific Proving Grounds) until 1962, ii) FSU in the Arctic, and iii) those conducted by France in Polynesia in the late 1960s and the early 1970s. The situation

89 is also characterised by the lack of significant fallout in the Southern hemisphere following the  
90 Chernobyl (1986) and Fukushima (2011) nuclear power plant accidents (Steinhauser et al., 2014).

91 The development of mass spectrometric techniques such as Inductively-Coupled Plasma  
92 Mass Spectrometry (ICP-MS) and Accelerator Mass Spectrometry (AMS) offered the opportunity  
93 to quantify ultra-trace levels of plutonium isotopes (fg to  $\mu\text{g g}^{-1}$ ) in a variety of contrasted  
94 environmental matrices (*e.g.* ice, soil, sediment, vegetation) after thorough chemical purifications.  
95 Importantly, ICP-MS and AMS provided powerful techniques to quantify individually  $^{240}\text{Pu}$  and  
96  $^{239}\text{Pu}$ , which was not possible in previous studies relying on alpha spectrometry, providing only  
97 the sum of both isotopes ( $^{239+240}\text{Pu}$ ).

98 Early records of radioactive fallout in the Southern hemisphere were derived from the  
99 analysis of ice cores collected in Antarctica (Koide et al., 1985). Although they were collected at  
100  $\sim 75^\circ$  latitude south ( $^\circ\text{S}$ ), *i.e.* in latitudinal bands far from those where France ( $22^\circ\text{S}$ ) or the USA  
101 (near the Equator) conducted detonations, their analysis demonstrated a change in the  $^{240}\text{Pu}/^{239}\text{Pu}$   
102 atom ratios with time. For example, the ratios found in ice deposited in the late 1950s dominated  
103 by the US tests in the Pacific Proving Ground (0.19–0.34) were higher than those accumulated in  
104 the early 1960s and dominated by the FSU tests (0.16–0.19), which indicates that Pu atom ratios  
105 provide relevant indicators to monitor potential changes in sources of nuclear fallout.

106 To address controversies (Arnaud et al., 2006; Guevara and Arribére, 2007; Magand and  
107 Arnaud, 2007) regarding the sources and the timing of radionuclide fallout at the scale of South  
108 America, our study provides a novel compilation of already published and new original  $^{240}\text{Pu}/^{239}\text{Pu}$   
109 atom ratio measurements conducted in soils and sediments collected across the subcontinent. As  
110 previously hypothesized (Chamizo et al., 2011; Kelley et al., 1999), South America was, in addition  
111 to global fallout following nuclear tests carried out in the Northern hemisphere, also partially  
112 exposed to the fallout associated with nuclear tests performed in the Southern hemisphere, notably  
113 the French nuclear tests conducted in Polynesia. To the best of our knowledge, this contribution  
114 had never been calculated so far and its consequences for various research fields in Earth Science  
115 (soil redistribution assessment, climatic and environmental reconstructions) would deserve the  
116 discussion that is provided in the current research, given the potential of these analyses to refine  
117 spatial and temporal interpretations derived from routine radionuclide measurements.

118

## 119 **2. Materials and Methods**

120

### 121 *2.1. Sample collection*

122 Detailed information regarding study site and sample collection are provided in the original  
123 publications listed in Table 1.

124 Sediment core was collected in 2020 in the Rincón del Bonete (RDB-01, 31 cm long) reservoir  
125 using a 63 mm Uwittec gravity corer equipped with a 1 meter PVC liner tube available at the  
126 Universidad de la República (Montevideo, Uruguay). Coring site was selected on the vicinity of  
127 the dams in order to ensure the maximum accumulation of fine sediment.

128 Another 83 cm long sediment core (CG02, S  $18^\circ 15.110'$ , W  $69^\circ 09.784'$ ) was collected in the  
129 western basin of Lake Chungará (Chile) in October 2014 at a water depth of 29.4 m, using a  
130 UWITEC gravity corer. Core opening and slicing were performed on the lake shore right after core  
131 collection to avoid transport-related disturbance. Detailed information for core sampling and  
132 processing are provided elsewhere (Guédron et al., 2019).

133           2.2. *Gamma spectrometry*

134           Prior to gamma spectrometry measurements, samples were dried at 40°C for at least 24 h,  
135 ground and passed through a 2 mm sieve, then placed in plastic Marinelli beakers. Gamma-ray  
136 measurements of <sup>137</sup>Cs (662 keV) in all samples were obtained using very low-background and  
137 high purity germanium detectors (HPGe) (Canberra) at Laboratoire des Sciences du Climat et de  
138 l'Environnement (LSCE, Gif-sur-Yvette, France) and the Laboratoire Souterrain de Modane  
139 (LSM, Modane, France). After quantification, <sup>137</sup>Cs activities (Bq kg<sup>-1</sup>) were decay-corrected to  
140 2020 assuming a decay constant of <sup>137</sup>Cs  $\lambda = \ln 2/30.2$  y.

141           The <sup>210</sup>Pb<sub>xs</sub> activities (T<sub>1/2</sub> = 22.3 years) were calculated by subtracting the supported activity  
142 (determined by using two <sup>226</sup>Ra daughters), <sup>214</sup>Pb activities (average count number at 295.2 and  
143 351.9 keV) and <sup>214</sup>Bi activity (609.3 keV) from the total <sup>210</sup>Pb activity measured at 46.5 keV.

144  
145           *Sample preparation and purification procedure for plutonium analysis*

146           The sample preparation and isotope composition measurements for samples collected in  
147 Lake Chungará and Rincón del Bonete reservoir were carried in the analytical expertise laboratory  
148 (Arpajon, France) of the Military Application Division (DAM) of the French Atomic Energy  
149 Commission (CEA). All acids used were of analytical grade and one or two process blanks were  
150 prepared under the same conditions as the samples. Dissolution, separation and purification of Pu  
151 followed a procedure almost similar than described in previous publications (Evrard et al., 2014;  
152 Jaegler et al., 2018). Quantities of approximately 5 g of each sample were transferred to Pyrex®  
153 beakers, covered with watch glasses to prevent cross contamination and dry-ashed at 450 °C for ~  
154 15 h in an electric furnace to decompose organic matter. After cooling to room temperature, limited  
155 amount of <sup>244</sup>Pu (~1 pg) was added as an isotopic dilution tracer for plutonium quantitative analysis.  
156 Digestion of the samples was carried out through five successive acid leaching steps: one leaching  
157 with concentrated HNO<sub>3</sub> (30 mL), one leaching with fuming HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> (1 mL during the  
158 second leaching), one leaching with concentrated HCl (30 mL) and a last one with 4 mol L<sup>-1</sup> HCl  
159 (50 mL). It should be noted that between each leaching step, the solutions were back flow heated  
160 at 120-150°C for several hours before being evaporated to dryness using hot plates. Samples were  
161 then filtrated with a disposable 0.45 µm Nalgene filtering unit (Thermo Scientific, Rochester, NY,  
162 USA). Dissolved fractions were evaporated to dryness before being recovered with 8 mol L<sup>-1</sup> HNO<sub>3</sub>  
163 (30 mL). Insoluble fractions were treated separately and transferred in clean Savillex® PFA  
164 Teflon® beakers, where a first digestion for 1 h with HF (20 mL) was done. Then, concentrated  
165 HNO<sub>3</sub> (5 mL) was added, followed by evaporation to dryness. Samples were recovered with HF (5  
166 mL) and concentrated HNO<sub>3</sub> (20 mL) and evaporated to dryness. Samples were finally recovered  
167 with concentrated HNO<sub>3</sub> (25 mL) and H<sub>2</sub>O<sub>2</sub> (1 mL), evaporated to dryness and recovered with 10  
168 mL of 8 mol L<sup>-1</sup> HNO<sub>3</sub>, before being added to the previous dissolved fractions and evaporated to  
169 dryness. Samples were recovered with 50 mL of 8 mol L<sup>-1</sup> HNO<sub>3</sub>, and a few mg of NH<sub>2</sub>OH.HCl  
170 and NaNO<sub>2</sub> were added to stabilize the (+IV) oxidation state of Pu and evaporated to dryness.  
171 Samples were finally recovered with 50 mL of 8 mol L<sup>-1</sup> HNO<sub>3</sub> before Pu separation on  
172 chromatographic columns.

173           Plutonium purification was performed with a 20 mL-column filled with Dowex AG1x8 anion-  
174 exchange resin (50-100 mesh AG1X8 and 100-200 mesh AG1X8 resins, 10 mL each), rinsed twice  
175 and conditioned with 60 mL of 8 mol L<sup>-1</sup> HNO<sub>3</sub>. A second purification was carried out with a 2  
176 mL-column filled with a Dowex AG1X4 anion-exchange resin (100-200 mesh resin). Samples  
177 were loaded in both columns, washed with 8 mol L<sup>-1</sup> HNO<sub>3</sub>, 10 mol L<sup>-1</sup> HCl and concentrated HCl

178 before Pu fraction elution with a  $\text{NH}_4\text{I}$  (1.5%)- 12 mol  $\text{L}^{-1}$   $\text{HCl}$  solution. A succession of  
179 evaporation to dryness and recovering was performed using concentrated  $\text{HNO}_3$ . Finally, samples  
180 were recovered with 3 mL of 2%  $\text{HNO}_3$  for ICP-MS measurements.

181 Sample preparation and isotope composition measurements for soils located in the  
182 Conceição River catchment (Brasil), in Antarctica and Peru were carried in the Laboratoire de  
183 Métrologie de la Radioactivité dans l'Environnement (LMRE) of the Institut de Radioprotection et  
184 de Sécurité Nucléaire (IRSN). Sample preparation was described in detail in previous papers and  
185 includes a two-step procedure allowing quantification of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  by alpha spectrometry,  
186 then separately  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  by SF-ICP-MS (Bouisset et al., 2021). Acids used were analytical  
187 grade for alpha spectrometry measurements, and superior quality for SF-ICP-MS. In summary,  
188 approximately 50g of samples were ashed and spiked with  $^{242}\text{Pu}$  (NIST SRM 4334). Digestion of  
189 the samples was carried out during 4 days using  $\text{HNO}_3$ ,  $\text{HCl}$  and  $\text{H}_2\text{O}_2$ , followed by three successive  
190 coprecipitations of actinides. Plutonium separation was then performed using chromatography  
191 column with anion exchange resin AG1X8 (Bio-rad), conditioned in 8 mol  $\text{L}^{-1}$   $\text{HNO}_3$ , and eluted  
192 with 12 mol  $\text{L}^{-1}$   $\text{HCl}/0.1$  mol  $\text{L}^{-1}$   $\text{NH}_4\text{I}$ . The purified fraction containing Pu was evaporated, before  
193 being dissolved in concentrated  $\text{HNO}_3$  and prepared by electrodeposition onto stainless steel discs  
194 (cathode) and Pt (anode). Pu isotopes were measured during 14 days with a low level background  
195 alpha spectrometer (alpha-analyst, Canberra) with 450  $\text{mm}^2$  PIPS detectors and using Genie 2000  
196 software (Canberra). Then, Pu electroplated samples were dissolved with concentrated  $\text{HNO}_3$  and  
197  $\text{HCl}$ . The solution was evaporated and recovered with 8 mol  $\text{L}^{-1}$   $\text{HNO}_3$ .  $\text{NaNO}_2$  was added for  
198 valence adjustment of Pu to Pu(IV). Plutonium purification was performed using a column filled  
199 with a Dowex AG1X8 anion-exchange resin and 8 mol  $\text{L}^{-1}$   $\text{HNO}_3$  before being eluted with  $\text{HCl}/\text{HI}$ .  
200 Finally, samples were evaporated to dryness and recovered with 0.5 mol  $\text{L}^{-1}$   $\text{HNO}_3$  for ICP-MS  
201 measurements.

202

### 203 *2.3. Isotope composition measurements by multiple-collector ICP-MS*

204 Pu isotopic composition for samples collected in Lake Chungará and Rincón del Bonete  
205 reservoir were measured with a Multi-Collector ICP-MS ("Neptune Plus", Thermo Fisher  
206 Scientific Inc., Bremen, Germany) equipped with eight Faraday cups and five ion counters. Ions  
207 counters were used for simultaneous measurements of plutonium isotopes ( $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  
208  $^{242}\text{Pu}$ ,  $^{244}\text{Pu}$ ). Faraday cups were used to monitor uranium content ( $^{238}\text{U}$ , to correct for  $^{238}\text{U}$  hydrides  
209 and peak tailing) and lead content (to correct for  $\text{PbO}_2$  interferences). An Aridus II (Cetac) as an  
210 introduction system, high efficiency sampler and skimmer cones, and a reinforced pumping in the  
211 interface were implemented to enhance the sensitivity (by approximately a factor of 10 with respect  
212 to the standard configuration). Two certified isotopic reference materials were used in ICP-MS  
213 measurements (SRM-947, SRM-948) for mass bias correction and plutonium isotopic verification.

214

215 For samples collected in Antarctica, Conceição (Brasil) and Peru, measurements were made  
216 with a sector field ICP-MS (SF-ICP-MS, Element 2, Thermo Fisher Scientific Inc., Bremen,  
217 Germany). The introduction system was made up of a PFA MicroFlow nebulizer (200  $\mu\text{L}/\text{min}$ )  
218 connected to a high sensitivity desolvating system (apex  $\Omega$ , ESI) and a cyclonic spray chamber  
219 (PC3 – ESI). A high capacity dry interface pump and a set of cones (Jet Interface) were used to  
220 enhance the sensitivity. Two reference material were used in alpha spectrometry and ICP-MS  
221 measurements (IAEA-410 – Bikini atoll sediment and IAEA-412 – Pacific Ocean sediment)

222

#### 223 2.4. Two-sources un-mixing model

224 The fraction of plutonium originating from a source with a much lower  $^{240}\text{Pu}/^{239}\text{Pu}$  atom  
225 ratio likely associated with French nuclear weapon tests (FNT) in the analysed samples was  
226 estimated as the first source in an empirical two-sources un-mixing. The second source being the  
227 global fallout (GF) associated with the pre-moratorium NWT. This model had been previously  
228 used to evaluate the contribution of fallout from two sources, where both GF and local/regional  
229 fallout have occurred on a given site (Kelley et al., 1999; Pittauer et al., 2017; Tighe et al., 2021).  
230 The compilation of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios published in the literature and the use of this two-  
231 sources un-mixing model was used to identify those latitudinal bands with atom ratios differing  
232 from the average global fallout signature.

233 Assuming that the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio determined in one sample ( $R_S$ ) is a mixture of both  
234 fallout sources, the first with a low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio ( $R_{\text{FNT}}$ ) and the second with a ratio  
235 corresponding to that of the global fallout ( $R_{\text{GF}}$ ):

$$236 R_S = \chi_{\text{FNT}} \times R_{\text{FNT}} + (1 - \chi_{\text{FNT}}) \times R_{\text{GF}} \quad (\text{Eq. 1})$$

237 where  $\chi_{\text{FNT}}$  and  $1 - \chi_{\text{FNT}}$  are respective contributions of FNT and GF. Consequently, the proportion  
238 of plutonium originating from this source with a much lower  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio, likely  
239 associated with FNT, is calculated according to Eq. 2:

$$240 \chi_{\text{FNT}} = \frac{R_S - R_{\text{GF}}}{R_{\text{FNT}} - R_{\text{GF}}} \quad (\text{Eq. 2})$$

241 The standard-uncertainty of the contribution  $u(\chi_{\text{FNT}})$  is determined by the combination of  
242 uncertainties associated with each variable in Eq. 2 and is expressed as follows:

$$243 u(\chi_{\text{FNT}}) = \chi_{\text{FNT}} \times \sqrt{\left(\frac{u(R_S)}{R_S - R_{\text{GF}}}\right)^2 + \left(\frac{u(R_{\text{FNT}})}{R_{\text{FNT}} - R_{\text{GF}}}\right)^2 + \left(\frac{(R_S - R_{\text{FNT}}) \times u(R_{\text{GF}})}{(R_S - R_{\text{GF}}) \times (R_{\text{FNT}} - R_{\text{GF}})}\right)^2} \quad (\text{Eq. 3})$$

244 The  $R_{\text{GF}}$  signatures were previously determined from a worldwide survey in terrestrial soils  
245 conducted between 1970-1971 by the Environmental Measurements Laboratory (EML) (Hardy et  
246 al., 1973; Krey et al., 1976) and analysed by TIMS (Thermal ionization mass spectrometry) for Pu  
247 atom ratio in the late 2000s (Fig. 2) (Kelley et al., 1999). Of note, radioactive fallout derived from  
248 the Chinese and French nuclear tests conducted after the sampling period could therefore not be  
249 recorded in these soil samples. In particular, approximately half of the tropospheric and  
250 stratospheric fallout associated with the FNT occurred after the sampling period. According to  
251 Kelley et al. (1999),  $R_{\text{GF}}$  is defined as  $0.178 \pm 0.019$ ,  $0.173 \pm 0.027$  and  $0.185 \pm 0.047$  for the 0-  
252  $10^\circ\text{N}$ ,  $0-30^\circ\text{S}$  and  $30-53^\circ\text{S}$  regions, respectively. Owing to the fact that uncertainties associated  
253 with these values take into account FNT (in particular the  $30-53^\circ\text{S}$  regions), we assume that a  $R_{\text{GF}}$   
254 signature of  $0.179 \pm 0.010$  is representative of the radionuclide fallout associated with the USA,  
255 FSU and UK tests at the scale of South America.

256 Most information published regarding  $R_{\text{FNT}}$  in tropospheric fallout is based on the analyses  
257 conducted on samples collected in 1996 by the International Atomic Energy Agency (IAEA),  
258 where a  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of  $0.04 \pm 0.01$  can be estimated for samples collected in Fangataufa  
259 atoll where part of the French tests was conducted (see the Discussion section). For  $R_S$  values

260 greater than  $R_{GF}$ , it is assumed that the Pu is fully derived from GF with corresponding  
261 contributions equal to 100%.

### 262 3. Results

263

#### 264 3.1. Spatial extent of plutonium signatures in soils

265 Uncertainties and/or standard deviations associated with Pu ratios presented in this section  
266 are detailed in Table 1.

267 The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios published in the literature and determined in the current  
268 research for four additional surface soil samples (0-5 cm) and two undisturbed soil profiles are  
269 displayed in Table 1 and in Fig. 2. Overall,  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios remain rather heterogeneous  
270 across South America, ranging from  $0.041 \pm 0.003$  to  $0.2045 \pm 0.0046$  with three distinct latitudinal  
271 patterns. For the equatorial latitudes ( $10^\circ\text{N}$ - $7^\circ\text{S}$ ),  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in surface soils and  
272 undisturbed soil profiles are characteristic of the average global fallout signature ( $\sim 0.18$ , Fig. 2).  
273 Although information is scarce for the Southern part of the continent (latitude below  $43^\circ\text{S}$ ), a  
274 similar pattern is observed in these regions although Pu atom ratios were found to be slightly higher  
275 in surface soils of Antarctica ( $0.19 \pm 0.01$ ), with a maximum  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of  $0.2045 \pm$   
276  $0.0046$  observed in a soil profile from Punta Arenas, Chile ( $53^\circ\text{S}$ , Fig. 2).

277 <Figure 2 here>

278 In contrast, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios found in soils located between  $20$  and  $42^\circ\text{S}$  on both  
279 sides of the Andean Cordillera significantly deviate from the average global fallout signature (Fig.  
280 2), with the lowest  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio ( $0.0041 \pm 0.003$ ) found at  $33^\circ\text{S}$  in a surface soil sample  
281 of La Parva (Chile) (Chamizo et al., 2011). From this location,  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios found in  
282 surface soils gradually increased southwards and northwards to reach the average global signature  
283 in Southern and Northern Chile. Compared to the signatures found in other soil profiles of South  
284 America, those obtained in Santiago, Chile, and Buenos Aires, Argentina, differ from the integrated  
285 fallout signature for the  $30$ - $53^\circ\text{S}$  regions ( $0.185 \pm 0.047$ ) (Kelley et al., 1999), with  $^{240}\text{Pu}/^{239}\text{Pu}$   
286 atom ratios of  $0.116 \pm 0.02$  and  $0.108 \pm 0.001$ , respectively (Table 1). In addition to the Buenos  
287 Aires sample collected on the Atlantic coast, the undisturbed soil profiles collected from the  
288 Conceição catchment ( $28^\circ\text{S}$ , Brazil) analysed in the current research showed  $^{240}\text{Pu}/^{239}\text{Pu}$  atom  
289 ratios remaining homogeneous with depth ( $0.135 \pm 0.006$ ), while being slightly lower than those  
290 found in the two soil profiles analysed by Kelley et al. (1999) in Rio de Janeiro ( $0.168 \pm 0.003$ )  
291 and Angra dos Reis ( $0.178 \pm 0.002$ ) located further North in Brazil ( $23^\circ\text{S}$ ).

292 <Table 1 here>

#### 293 3.2. Respective contributions of global vs. French fallout (FF) in soils

294 The proportion of plutonium originating from the FNT, calculated from the two-source un-  
295 mixing model (see section 2.5.), is provided in Table 1 and displayed in Fig. 3. Accordingly, the  
296 highest contributions of FNT were calculated for surface soil samples collected on the western  
297 continental margin of South America, in La Parva ( $99.3 \pm 7.5\%$ ,  $33^\circ\text{S}$ ) and Chillan ( $48.8 \pm 7.7\%$ ,  
298  $36^\circ\text{S}$ ) both located in Chile (Fig. 3). In undisturbed soil profiles, the proportion increased between  
299  $23^\circ\text{S}$  to  $35^\circ\text{S}$  from  $8.0 \pm 6.8\%$  in Rio de Janeiro, Brazil, to  $51.3 \pm 5.1\%$  in Buenos Aires, Argentina.  
300 On the contrary, the contribution of GF remained unambiguously the main source of Pu found in



301 soils located in the Northern and Southern parts of the subcontinent, with proportions generally  
302 exceeding 80% (Fig. 3).

303 <Figure 3 here>

### 304 3.3. Temporal evolution of fallout signature in sediment core profiles

305 The  $^{210}\text{Pb}_{\text{xs}}$  dating method was used to provide the chronology of two sedimentary  
306 sequences collected in Lake Chungará (Chile, 18°S – 69°W) and in the Rincón del Bonete  
307 Reservoir (Uruguay, 32°S – 56°W). The Constant Flux Constant Sedimentation (CF:CS) was  
308 applied to determine  $^{210}\text{Pb}_{\text{xs}}$  age models for these sediment cores (Bruel and Sabatier, 2020;  
309 Goldberg, 1963; Krishnaswamy et al., 1971) (Table 2).

310 <Table 2 here>

311 For Lake Chungará, a mean sedimentation rate of  $0.95 \pm 0.04 \text{ mm yr}^{-1}$  ( $1\sigma$ ) over the top 10  
312 cm was previously determined (Guédron et al., 2019). Samples from successive layers had to be  
313 regrouped as there was not sufficient material to perform high temporal-resolution analyses.  
314 Plutonium isotopes were measured for three samples covering the periods 1908-1955 (5.75 – 9.75  
315 cm), 1955-1987 (2.75-5.25 cm) and 1987-2014 (2.75-0 cm) according to the CF:CS model (see  
316 Supplementary Fig. S1). The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio gradually decreased with time from  $0.252 \pm$   
317  $0.015$  (1908-1955) to  $0.127 \pm 0.013$  between 1987-2014 (proportion from FNT,  $37.3 \pm 10.7\%$ ).  
318 Sample CG6-11 corresponding to sediment deposited between 1955-1987, where the maximum  
319 activity peaks of  $^{137}\text{Cs}$  were detected, showed a  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of  $0.156 \pm 0.009$  (FNT:  $16.3$   
320  $\pm 8.9\%$ ) (Fig. 4).

321 <Figure 4 here>

322 Higher down-core resolution with depth was available for the sediment core collected in  
323 the Rincón del Bonete Reservoir, in Uruguay (Fig. 5). Importantly, sampling reached the bedrock  
324 material and it was therefore assumed to cover the entire sedimentation period since the reservoir  
325 has become operational early in the 1950s. In contrast, the CF:CS model applied to the  $^{210}\text{Pb}_{\text{xs}}$   
326 activities calculated a mean accumulation rate of  $11.03 \pm 1.31 \text{ mm yr}^{-1}$  ( $1\sigma$ ) over the uppermost 30  
327 cm, which indicated that this sediment was deposited since  $1991 \pm 4$  years onwards (see Fig. S2).  
328 The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios showed the occurrence of uniform values with depth, ranging from  
329 0.082 to 0.107 with an average signature of  $0.092 \pm 0.015$ , indicating a mean contribution of  $62.2$   
330  $\pm 12.2\%$  from FNT in this sediment core.

331 <Figure 5 here>

332

## 333 4. Discussion

334

### 335 4.1. Signatures and spatial extent of French radioactive debris

336 The complete cessation of nuclear weapon testing by France took place in January 1996  
337 following a final series of six underground tests conducted in Polynesia. The IAEA was mandated  
338 on the same year to assess the radiological situation at the C.E.P (IAEA, 1998). Approximately 300

339 samples were collected in contrasted matrices (e.g. corals, soil profiles, aerosols, etc.) and analysed  
340 for their content in  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ . Based on this survey, it was concluded that most of  
341 the residual radioactivity in both atolls was derived from safety trials and barge tests, which have  
342 led to significant local fallout. Then, in 2005, part of these samples was analysed by means of  
343 AMS, and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios characteristic of low yield detonations were observed (Hrnecek  
344 et al., 2005) (see Table S1). In safety trial sites of Moruroa, a  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of 0.019 was  
345 determined, in close agreement with the signatures lower than 0.03 measured by Chiappini et al.  
346 (1999) in lagoon sediments. In other areas of Moruroa atoll affected by detonations conducted on  
347 barges,  $^{240}\text{Pu}/^{239}\text{Pu}$  atoms ratios were found to be slightly higher and close to 0.05 in topsoils and  
348 loose coral rocks (Chiappini et al., 1999; Hrnecek et al., 2005). For samples collected at Fangataufa  
349 atoll, similar low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio signatures of  $\sim 0.05$  associated with the Rigel barge tests  
350 (125 kT) conducted in 1966 were outlined in these studies. Recently, the contribution of close-in  
351 fallout to the Pu deposition was investigated in Gambier Islands located 400 km south-east from  
352 the C.E.P. The authors found an average  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios of  $0.0394 \pm 0.0062$  in seven  
353 undisturbed soil profiles (Bouisset et al., 2021).

354 With the exception of four devices detonated on barges and five safety trials conducted at  
355 the soil surface, 34 atmospheric tests were carried out from balloons tethered at an altitude of  
356 several hundred meters above the atolls. Of note, this type of device has been used to limit both  
357 local and regional fallout in Polynesian regions, thereby injecting large proportions of fission  
358 products into the upper troposphere or stratosphere depending on the yield. Despite the lack of  
359 public information regarding individual signatures for these tests conducted in altitude, the  
360 available data suggests that the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios associated with the original low-yield  
361 devices remained in the same range (0.03 – 0.07) as other weapon grade devices, such as those  
362 detonated in Australia, Nevada Test Site and in the Pacific Proving Grounds by the UK and the  
363 U.S (Froehlich et al., 2016; Hamilton et al., 2009; Hicks and Barr, 1984; Muramatsu et al., 2001;  
364 Tims et al., 2013). In the light of the publicly available information, the  $R_{\text{FNT}}$  of  $0.04 \pm 0.01$  used  
365 in the un-mixing model previously described appears to be the most relevant to date.

366 Owing that a large proportion of FNT were conducted during the southern winter when a  
367 dominant general circulation of air masses at mid-latitudes is characterised by West to East  
368 directions (Cai et al., 2020), most of the radioactive particles injected into the troposphere  
369 following kiloton explosions were likely first deposited in South America during the subsequent  
370 days or weeks. This hypothesis is further supported by a set of declassified documents and maps  
371 published by the French Ministry of Defence in 2013 regarding the spatial extent and trajectories  
372 of radioactive clouds released from nuclear tests conducted by France in 1967 and 1968. In  
373 addition, the low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios observed in surface soils from La Parva (Chile) in  
374 particular (ranging from 0.024 to 0.067, average  $0.041 \pm 0.003$ ) (Chamizo et al., 2011), and in soils  
375 located at similar latitudes as the C.E.P in general, strongly support the occurrence of direct  
376 tropospheric fallout of FNT debris in these regions of South America.

377 In contrast, the contribution of stratospheric fallout from high-yield FNT to the Pu atom  
378 ratios in soils cannot be estimated based on the current information available in the literature.  
379 Information regarding the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios signatures from stratospheric injections is more  
380 limited and based on the analysis of air filters collected by the Health and Safety Laboratory  
381 (HASL) from 1963 to 1970 at  $40^{\circ}\text{S}$ ,  $70^{\circ}\text{W}$  between 10 and 16 km of altitude (Fig. 6). Similarly to  
382 what can be observed in sediment cores analysed in the current research, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios

383 decreased from a GF signature ( $0.181 \pm 0.005$ ,  $1\sigma$ ) between 1963 - 1966 to a lower signature ( $0.11$   
384  $\pm 0.02$ ,  $1\sigma$ ) in 1969-1970, which is in good agreement with those values reported for soils and  
385 sediment cores collected at similar latitudes.

#### 386 *4.2. Impact of plutonium signatures to improve sediment dating.*

387 Although not assessed in the current study, potential  $^{137}\text{Cs}$  and plutonium mobility in  
388 sediment cores has to be considered through additional analyses, such as grain size, mineralogy  
389 and organic matter properties.

390 A plutonium isotopic signature characteristic of the high yield atmospheric tests conducted  
391 in the 1950s by the US in the Pacific Proving Ground (Fig. 1) is observed in the lower part of the  
392 Chungará sediment core with  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of  $0.252 \pm 0.015$  in sample CG12-20  
393 (Muramatsu et al., 2001). According to the  $^{210}\text{Pb}_{\text{xs}}$  age model, sample CG6-11 corresponds to  
394 sediment deposited between 1955 and 1987, thereby reflecting the main radioactive fallout period  
395 and containing fallout originating from tests conducted in both the Northern and Southern  
396 hemispheres. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of  $0.156 \pm 0.009$  determined in this sample is in perfect  
397 agreement with the signature proposed by Bouisset et al. (2021) for the Southern hemisphere ( $0.157$   
398  $\pm 0.011$ ), including fallout from all the Nuclear Weapon States. A similar value was observed in  
399 an undisturbed soil profile collected by the EML in Lima, Peru ( $0.1565$ ,  $12^\circ\text{S}$ ), suggesting that  
400 FNT debris, although they are less abundant, can also be observed in the Northern part of South  
401 America (Fig. 3). The upward circulation that occurs in the equatorial regions (known as Hardley  
402 circulation) has likely limited the deposition of FNT debris in these zones, as suggested by the  
403  $^{240}\text{Pu}/^{239}\text{Pu}$  signature of  $\sim 0.18$  determined in French Guiana and soil surface samples collected in  
404 Peru in the framework of the current research. In contrast, the decreasing atom ratio observed in  
405 sample CG1-5 ( $0.127 \pm 0.013$ ) for Lake Chungará sediments deposited between 1987-2014 is very  
406 likely related to the higher proportion of FNT debris.

407 The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in sediment deposited in the Rincón del Bonete Reservoir,  
408 although they are significantly lower than those measured in Lake Chungará, remained in  
409 agreement with the plutonium signatures reported for undisturbed soil profiles collected at similar  
410 latitudes by Kelley et al. (1999) ( $0.1156 \pm 0.0017$  and  $0.1075 \pm 0.0007$  in Santiago and Buenos  
411 Aires, respectively). The CF:CS  $^{210}\text{Pb}_{\text{xs}}$  age model for this sediment core indicates sediment  
412 accumulation since 1990. The analysis of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in this lacustrine archive  
413 confirmed that the sampled sequence covered the most recent period (post-1990) when land use  
414 change has dramatically modified sediment dynamics in this river catchment, with a brutal  
415 transition from “pristine” natural grassland conditions to their conversion into cropland.  
416 Considering the absence of a Pu signature characteristic of the 1964-1965 fallout peak, we  
417 demonstrate the added value of analysing  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in addition to  $^{137}\text{Cs}$  and/or  $^{210}\text{Pb}_{\text{xs}}$   
418 in sediment profiles collected in this region of South America. This will avoid any age model  
419 misinterpretation that may arise from an incorrect sediment dating from short-lived radionuclides  
420 (see supplementary Fig. 2) or the lack of other local knowledge (e.g., regional land use changes).

#### 421 *4.3. Wider implications*

422 South America has been exposed to significant land use change since the 1960s – a period  
423 that coincides with that of the main radioactive fallout (Winkler et al., 2021). The conversion of  
424 natural forests and grassland into cropland may result in the degradation of terrestrial ecosystems  
425 and to an excessive sediment supply to lacustrine environments and river systems, as observed in

426 the Rincón del Bonete Reservoir. Accordingly, improving the chronology of this reconstruction is  
427 of utmost interest in this part of the world. The current research demonstrates that the analysis of  
428  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in addition to that of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  in lacustrine archives may contribute to  
429 this ultimate objective given the strong spatial and temporal variations of this ratio compiled across  
430 the continent and demonstrated in this study.

431 Low levels of  $^{137}\text{Cs}$  analysed in soils and sediment of the Southern hemisphere outline the  
432 need to find alternative chronomarkers to reconstruct soil redistribution rates and/or to cross-check  
433 and validate the dating of sediment archives. Pu is a powerful candidate to achieve this goal as it  
434 may allow further distinguishing between different time periods corresponding to contrasted  
435 nuclear testing stages or fallout originating from nuclear experimentations carried out by different  
436 countries based on the reconstruction of  $^{240}\text{Pu}/^{239}\text{Pu}$  temporal evolution in sediment cores. The  
437  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio should therefore be analysed in sediment core layers showing a single or  
438 multiple  $^{137}\text{Cs}$  fallout peaks to avoid any misinterpretation in the core dating and, consequently, on  
439 the chronology of the reconstruction of environmental processes that have occurred in this  
440 subcontinent in general, and in the central part of South America in particular.

441 Models converting  $^{137}\text{Cs}$  measurements (Mabit et al., 2014) into quantitative estimates of  
442 erosional and depositional rates over the past decades are based on the assumption that radionuclide  
443 fallout peaked in 1963. Based on the undisturbed soil profiles of Conceição, Brazil, we showed  
444 that 30% of Pu deposition could be attributed to FNT at this site. Recalibrating these soil erosion  
445 models for applications in mid-latitudes of South America (by shifting the fallout peak to the period  
446 1965-1970 instead of 1963 as it is currently done) would better constrain soil redistribution rates  
447 calculated in these regions.

448 In addition to the analysis of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios,  $^{238}\text{Pu}$  ( $T_{1/2} = 88$  years) fallout peak in  
449 1968, resulting from the atmospheric disintegration of a nuclear auxiliary power generator in 1964  
450 (SNAP9-A), may provide an additional time marker (Hancock et al., 2011) to confirm the  
451 occurrence of a radioactive fallout peak in 1970 in some regions of South America as suggested in  
452 our study.

453 Accordingly, the current research encourages the scientific community to continue  
454 analysing Pu in general, and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in particular, to improve our understanding of  
455 the spatial distribution of Pu fallout and its chronology in the Southern Hemisphere. This improved  
456 knowledge will undoubtedly provide a powerful anthropogenic marker that will contribute refining  
457 the dating of sediment cores, with a focus on those processes that took place during the 1960s and  
458 the 1970s when the conversion of natural areas into agricultural land increased around the world  
459 in general, and in Southern America in particular.

## 460 461 **5. Conclusions**

462 A novel compilation of already published and new original plutonium isotopes ( $^{239,240}\text{Pu}$ )  
463 measurements conducted in lake sediments and soils allowed us to update our knowledge of the  
464 spatial and temporal distribution of artificial radioactive fallout in South America. Significantly  
465 lower  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios differing from the average global fallout signature were found in the  
466 20-45° latitudinal band and attributed to the higher contribution of the French atmospheric nuclear  
467 weapon tests (1966-1974) to the deposition of plutonium in these soils and sediments. These results  
468 indicate that fallout radionuclides from French atmospheric tests should be considered to avoid any

469 misinterpretation in sediment core dating and to refine the chronology of the reconstruction of  
470 environmental processes in this region of the world.

471

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481

482

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484

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487

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489 S.M., S.G., P.C., M.C. and G.C. collected the samples. P.A.C., O.E., F.P., P.S., A.F., T.T.,  
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492 P.S., M.T. and M.C. performed the analysis and measurements (radiochemistry, gamma  
493 spectrometry and ICP-MS measurements). All authors revised the manuscript to prepare  
494 the final version.

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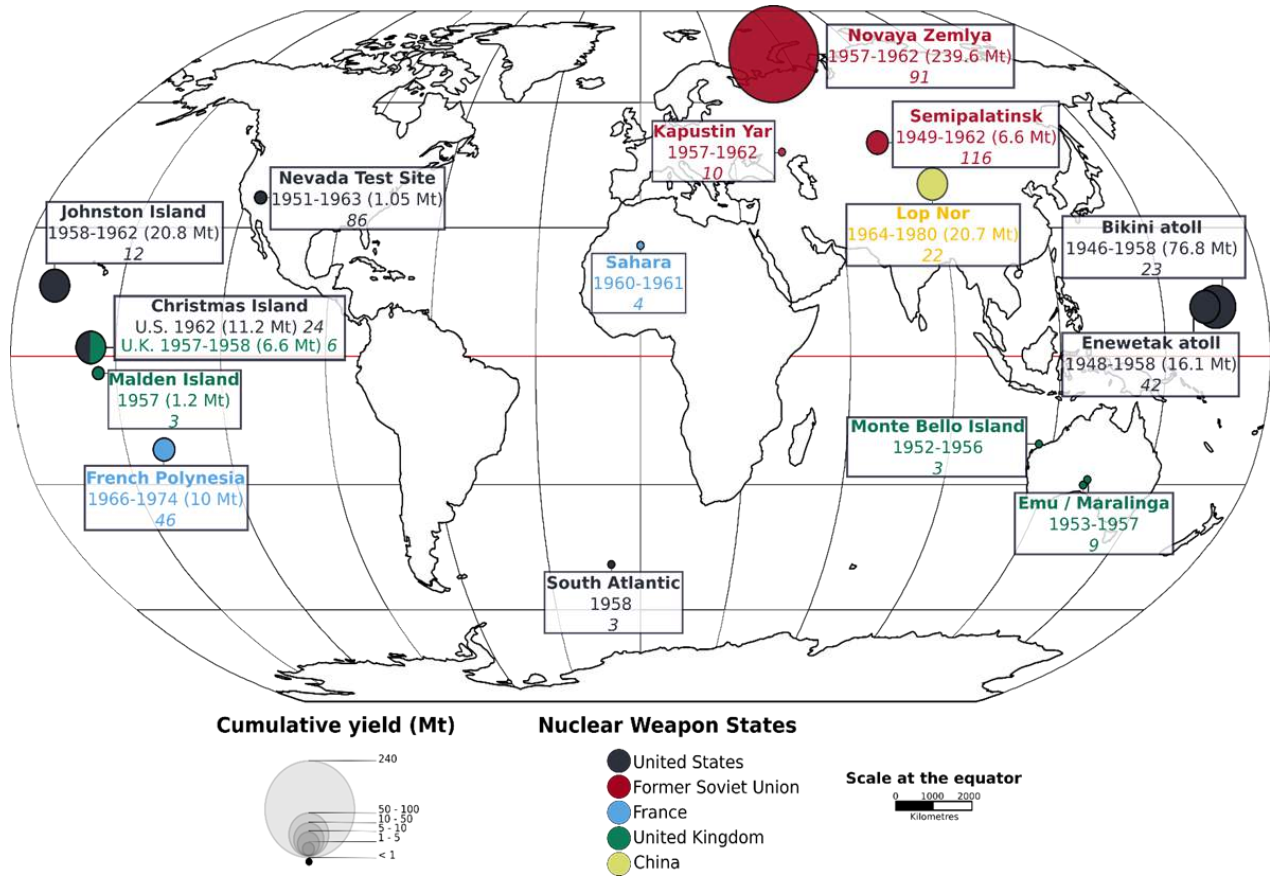
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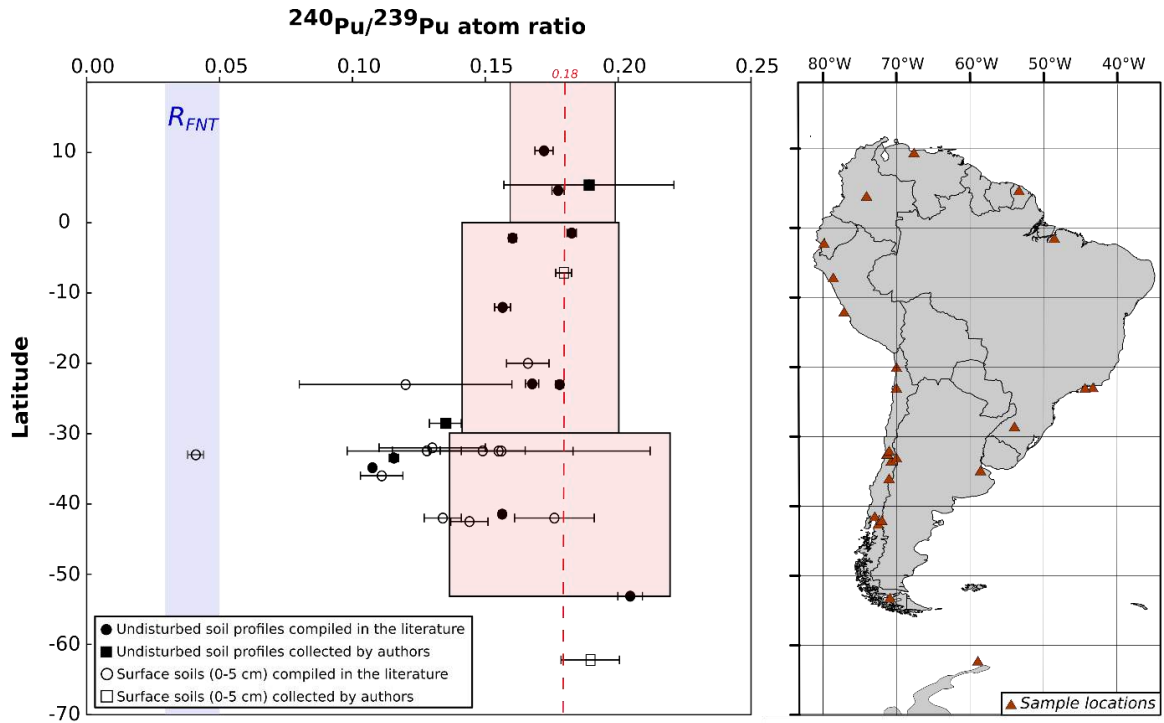
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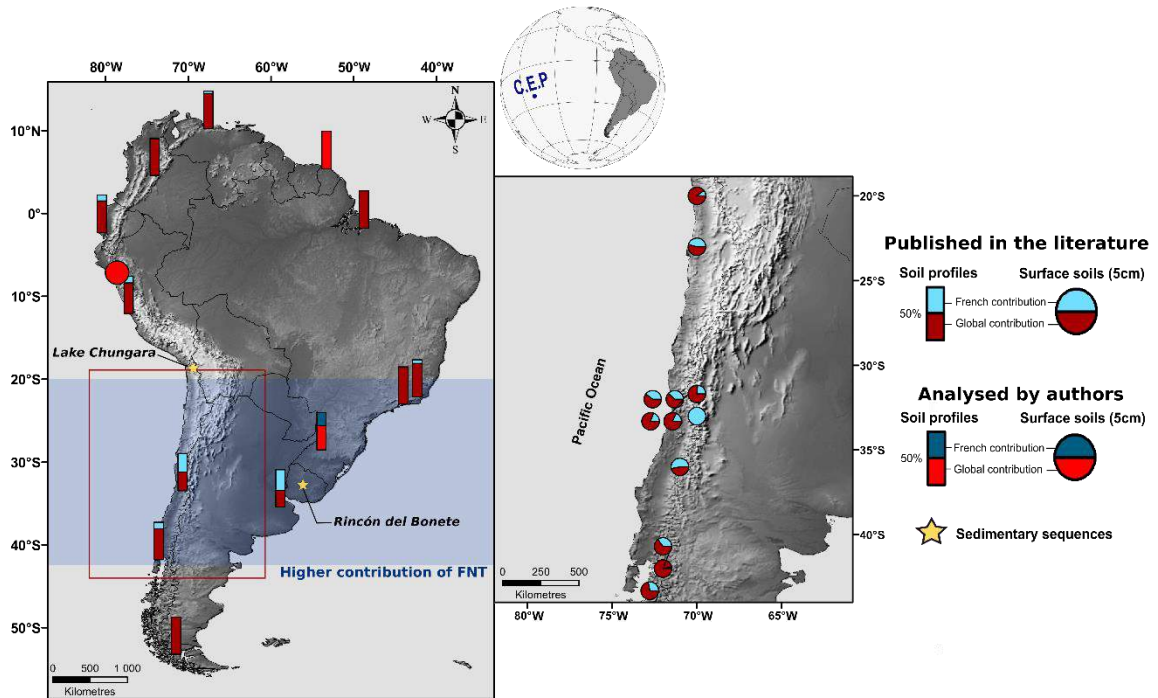




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 658 **Fig. 1.** Location, timing, number (in italics) and total yield of atmospheric nuclear tests conducted  
 659 around the world (8). Two operational nuclear detonations (Hiroshima, Nagasaki), 39 safety  
 660 trials, four tests in the Pacific Ocean, one in New Mexico (USA) and two tests in Aralsk  
 661 (Former Soviet Union) with a cumulative yield of approximately 0.2 Mt (0.045% of the  
 662 total yield) are not shown for clarity.

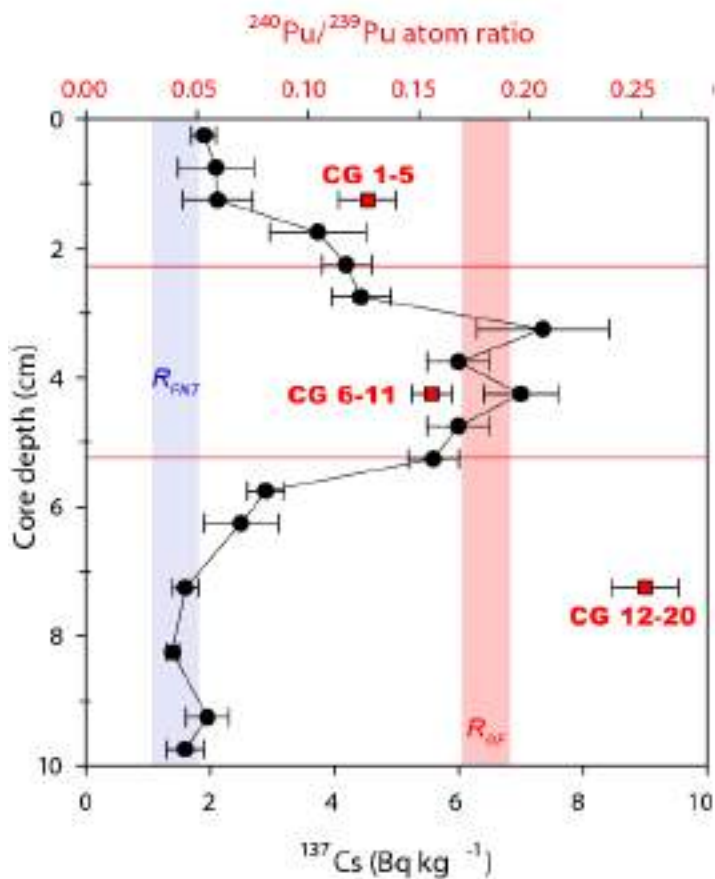


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 665 **Fig. 2.** Latitudinal distribution of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in undisturbed soil profiles and surface  
 666 soils across continental South America and Antarctica published in the literature (circle)  
 667 and analysed in the current research (square). The red rectangles indicate the integrated  
 668 fallout signatures in different latitudinal ranges as determined from soils collected in 1970-  
 669 1971 (19), while the blue rectangle ( $R_{FNT}$ ) indicates the French fallout signature reported in  
 670 previous studies. The average global fallout signature in soils ( $\sim 0.18$ ) is displayed as a red  
 671 dashed line.  
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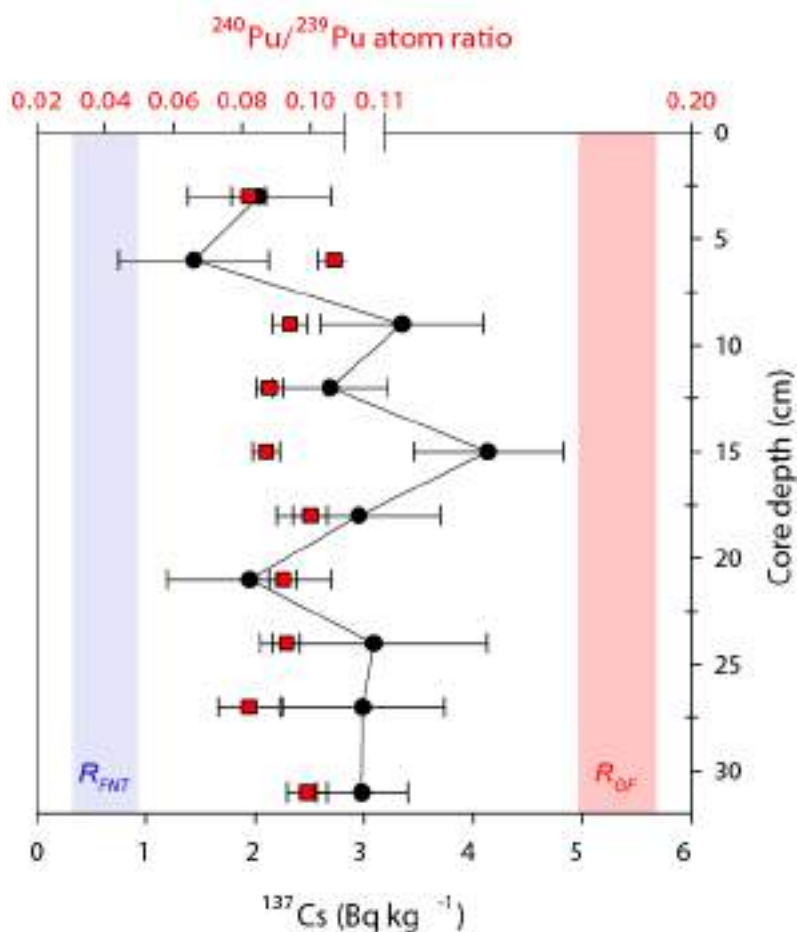
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 674 **Fig. 3.** Respective contributions of global and French fallout in South America surface soil samples  
 675 and undisturbed soil profiles published in the literature and analysed in the current research.  
 676 Contribution uncertainties for each sample can be found in Table 1. Soil samples from  
 677 Antarctica are not displayed for clarity. The location of the *Centre pour l'Expérimentation*  
 678 *du Pacifique* (C.E.P.) is shown in the inset world map.  
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Lake Chungara (18°S)

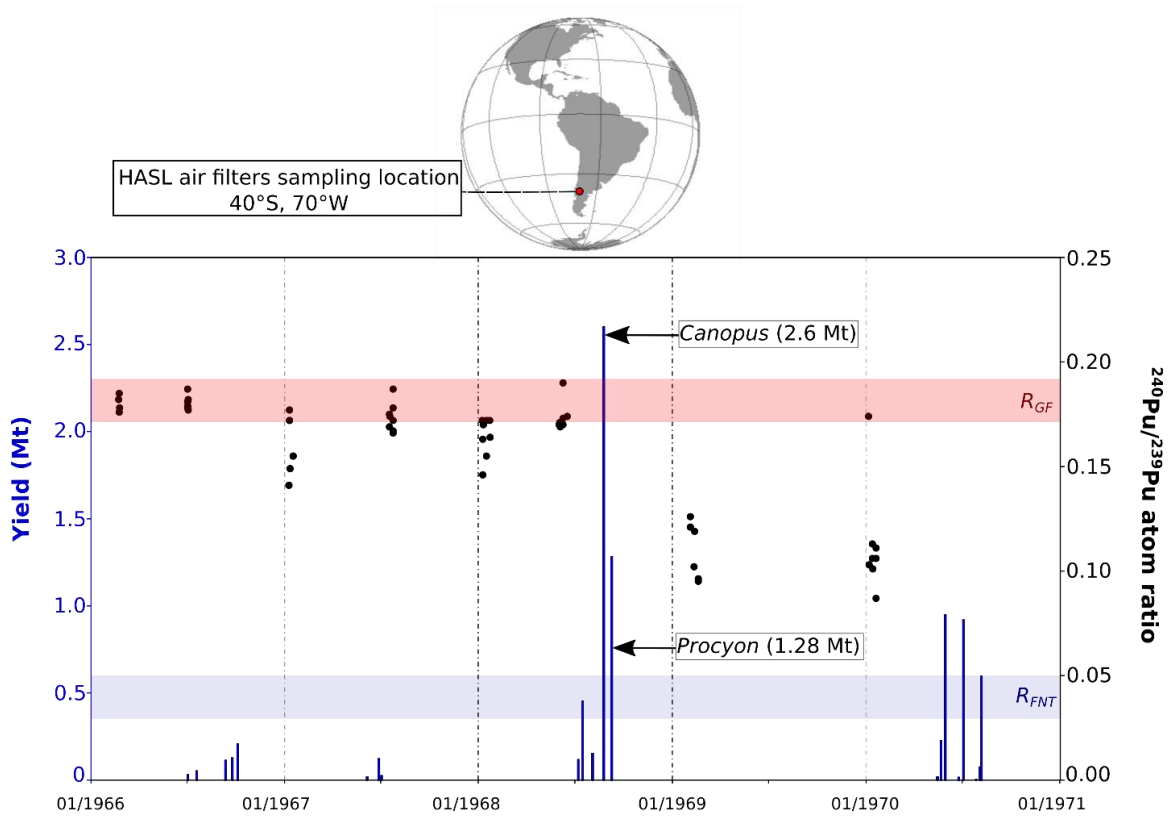


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 681 **Fig. 4.** Temporal evolution of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios (red squares) and  $^{137}\text{Cs}$  activities (black dots)  
 682 in sediment core collected in Lake Chungara (Chile). The French ( $R_{\text{FNT}}$ , in blue) and global  
 683 ( $R_{\text{GF}}$ , in red)  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio fallout signatures used in the current research are  
 684 provided as an indication.  
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### Rincón del Bonete (32°S)



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 688 **Fig. 5:** Temporal evolution of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios (red squares) and  $^{137}\text{Cs}$  activities (black dots)  
 689 in sediment core collected in Rincón del Bonete reservoir (Uruguay). The French ( $R_{\text{FNT}}$ , in blue)  
 690 and global ( $R_{\text{GF}}$ , in red)  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio fallout signatures used in the current  
 691 research are provided as an indication. Please note the scale difference for reporting the  
 692  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in the sediment core.  
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 695 **Fig. 6.**  $^{240}\text{Pu}/^{239}\text{Pu}$  temporal evolution in air filters collected by HASL between 1965 and 1971 at  
 696  $40^\circ\text{S}$  latitude (black dots, no uncertainties for these samples were provided in the initial  
 697 database), plotted along with French nuclear test yields over the same period. The  $R_{\text{FNT}}$  (in  
 698 blue) and  $R_{\text{GF}}$  (in red) fallout signatures used in the current research are provided as an  
 699 indication  
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**Table 1.** <sup>240</sup>Pu/<sup>239</sup>Pu atom ratios in soils of South America published in the literature and analysed in the current research. <sup>a</sup>Sampling carried out in other studies and analysed for Pu isotopes by the authors; <sup>b</sup><sup>240</sup>Pu/<sup>239</sup>Pu atom ratios as reported in Table 2 from Kelley et al., 1999; <sup>c</sup>"Weighted mean of duplicate aliquots from the same sample collection" (19); \* Samples analysed at the IRSN; \*\* Samples analysed at the CEA/DAM.

Country	Latitude	Longitude	Study Area / Sample label	Elevation (m)	Sampling year	<sup>240</sup> Pu/ <sup>239</sup> Pu	Proportion of Plutonium from French fallout	Measurement technique	Author	Note
French Guiana**	5.41	-53.31	ROC	84	2016	0.189 ± 0.032	0%	ICP-MS	de Tombeur et al., 2020 <sup>a</sup>	Undisturbed soil profile (0-40 cm) - Measurement ± 2σ error
Peru*	-7.13	-78.61	PASO02	3630	2017	0.178 ± 0.013	0.5 ± 11.7%	ICP-MS	This study	Surface soil (0-5cm)- Measurement ± 2σ error
Peru*	-7.14	-78.60	PASO05	3570	2017	0.181 ± 0.015	0%		This study	Surface soil (0-5cm)- Measurement ± 2σ error
Brazil*	-28.53	-53.92	CON 2-4cm	270	01/2016	0.132 ± 0.011	33.7 ± 9.5%	ICP-MS	Didoné et al., 2019 <sup>a</sup>	Undisturbed soil profile (0-42 cm) Measurement ± 2σ error
			CON 6-8cm			0.131 ± 0.011	34.4 ± 9.5%			
			CON 10-12cm			0.137 ± 0.012	30 ± 10.2%			
			CON 14-16cm			0.133 ± 0.011	32.9 ± 9.6%			
			CON 18-20cm			0.14 ± 0.016	27.9 ± 12.8%			
			Conceição average	/	/	0.135 ± 0.006	31.5 ± 6.9%			<sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 2σ (n=5)
Antarctica*	-62.19	-58.91	A1	17	2017	0.195 ± 0.015	0%	ICP-MS	This study	Surface soil (0-5cm)- Measurement ± 2σ error
	-62.19	-58.91	A2	17	2017	0.184 ± 0.018	0%			This study
<b>Published in the literature</b>										
Venezuela	10.25	-67.6	Maracay S-1598	n.d.	10/1970 to 01/1971	0.1721 ± 0.0034	4.7 ± 7.2%	TIMS	Kelley et al., 1999 <sup>b</sup>	Undisturbed area sampled to a depth of 30cm Measurement ± 1σ error
Colombia	4.6	-74.08	Bogota S-1599	n.d.	10/1970 to 01/1971	0.1774 ± 0.00232 <sup>c</sup>	0.9 ± 7.2%			
Brazil	-1.45	-48.48	Belem S-1607	n.d.	10/1970 to 01/1971	0.1825 ± 0.00176	n.d.			
Equador	-2.17	-79.87	Guayaquil S-1627	n.d.	10/1970 to 01/1971	0.1602 ± 0.00151 <sup>c</sup>	13.3 ± 6.3%			
Peru	-12.02	-77.13	Lima S-1632	n.d.	10/1970 to 01/1971	0.1565 ± 0.003 <sup>c</sup>	16.0 ± 6.4%			
Brazil	-22.9	-43.23	Rio de Janeiro S-1728/9	n.d.	10/1970 to 01/1971	0.1676 ± 0.0025	8.0 ± 6.8%			
Brazil	-23	-44.3	Angra dos Reis S-1605	n.d.	10/1970 to 01/1971	0.178 ± 0.0015	0.5 ± 7.1%			
Chile	-33.45	-70.7	Santiago S-1708	n.d.	10/1970 to 01/1971	0.1156 ± 0.0017	45.5 ± 5.2%			
Argentina	-34.85	-58.53	Buenos Aires S-1642	n.d.	10/1970 to 01/1971	0.1075 ± 0.0007 <sup>c</sup>	51.3 ± 5.1%			
Chile	-41.45	-72.95	Puerto Montt S-1690	n.d.	10/1970 to 01/1971	0.1563 ± 0.0013 <sup>c</sup>	16.1 ± 6.1%			
Chile	-53.13	-70.88	Punta Arenas S-1707	n.d.	10/1970 to 01/1971	0.2045 ± 0.0046	0%			
Chile	-32.43	-71.24	La Greda	n.d.	From 2009 to 2010	0.149 ± 0.016	21.4 ± 12.9%	AMS	Salmani-Ghabeshi et al., 2018	Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=4)
Chile	-32.46	-71.27	Los Maitenes	n.d.	From 2007 to 2010	0.155 ± 0.057	17.1 ± 41.5%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-32.45	-71.28	Puchuncavi	n.d.	From 2007 to 2010	0.128 ± 0.013	36.5 ± 10.7%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-32.48	-71.26	Valle Alegre	n.d.	From 2009 to 2010	0.156 ± 0.027	16.3 ± 20.4%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-42.51	-72.47	Santa Barbara	n.d.	2009	0.144 ± 0.007	25.0 ± 7.6%			Surface soil (0-5cm)- Measurement ± 1σ
Chile	-20	-70	Iquique	Sea level	From 2007 to 2009	0.166 ± 0.008	9.1 ± 8.7%	AMS	Chamizo et al., 2011	Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-23	-70	Antofagasta	Sea level		0.12 ± 0.04	42.3 ± 29.3%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-32	-71	Punchuncavi	Sea level		0.13 ± 0.02	35.1 ± 15.4%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-33	-70	La Parva	2800		0.041 ± 0.003	99.3 ± 7.5%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-36	-71	Chillan	Sea level		0.111 ± 0.008	48.8 ± 7.7%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-42	-72	Santa Barbara	Sea level		0.134 ± 0.007	32.2 ± 7.4%			Surface soil (0-5cm) - <sup>240</sup> Pu/ <sup>239</sup> Pu = Average ± 1σ (n=3)
Chile	-42	-72	Chaiten	1280		0.176 ± 0.015	1.9 ± 12.9%			Surface soil (0-5cm)- Measurement ± 1σ

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**Table 2.**  $^{210}\text{Pb}_{\text{xs}}$  data, CF:CS derived ages and Temporal evolution of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios for the Lake Chungara and Rincón del Bonete reservoir sediment cores. <sup>a</sup>Analytical measurement uncertainty (k=2). <sup>b</sup>For Chungara lake,  $^{137}\text{Cs}$  analyses were not carried out for samples which have been grouped. <sup>c</sup>For Chungara,  $^{137}\text{Cs}$  activities are expressed from the time of collection (2014).

Sediment core	Samples	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{239+240}\text{Pu}$ concentration (fg g <sup>-1</sup> ) <sup>a</sup>	Proportion of Plutonium from French fallout	Depth Interval (cm)	$^{137}\text{Cs}$ (Bq kg <sup>-1</sup> , 2020) <sup>abc</sup>	Calendar Age (year) - CF:CS		
							Age	Minimum Age	Maximum Age
Lake Chungara	CG 1-5	0.127 ± 0.013	31.5 ± 1.9	37.3 ± 10.7%	0.25	1.9 ± 0.2	2011	2011	2011
					0.75	2.1 ± 0.6	2006	2006	2006
					1.25	2.1 ± 0.6	2001	2001	2000
					1.75	3.7 ± 0.8	1996	1996	1995
					2.25	4.2 ± 0.4	1990	1991	1990
	CG 6-11	0.156 ± 0.009	214.9 ± 7.3	16.3 ± 8.9%	2.75	4.4 ± 0.5	1985	1986	1984
					3.25	7.4 ± 1.1	1980	1981	1979
					3.75	6.0 ± 0.5	1974	1976	1973
					4.25	7.0 ± 0.6	1969	1970	1968
					4.75	6.0 ± 0.5	1964	1965	1962
	CG 12-20	0.252 ± 0.015	39.8 ± 1.5	n.d.	5.25	5.6 ± 0.4	1959	1960	1957
					5.75	2.9 ± 0.3	1953	1955	1951
					6.25	2.5 ± 0.6	1948	1950	1946
					7.25	1.6 ± 0.2	1938	1940	1935
					8.25	1.4 ± 0.1	1927	1929	1924
Rincon del Bonete Reservoir	BON1	0.082 ± 0.005	65.3 ± 1.6	69.7 ± 6.5%	3	2.0 ± 0.7	2017	2016	2017
	BON2	0.107 ± 0.005	46.9 ± 1.3	51.7 ± 6.2%	6	1.4 ± 0.7	2014	2013	2015
	BON3	0.094 ± 0.005	54.7 ± 1.3	61.1 ± 6.3%	9	3.3 ± 0.7	2011	2010	2012
	BON4	0.088 ± 0.004	53.8 ± 1.2	65.4 ± 6.0%	12	2.7 ± 0.5	2009	2007	2010
	BON5	0.087 ± 0.004	59.0 ± 1.8	66.1 ± 6.1%	15	4.1 ± 0.7	2006	2004	2008
	BON6	0.100 ± 0.005	51.4 ± 1.7	56.7 ± 6.3%	18	2.9 ± 0.7	2003	2001	2005
	BON7	0.092 ± 0.004	54.9 ± 1.7	62.5 ± 6.0%	21	1.9 ± 0.7	2000	1998	2003
	BON8	0.093 ± 0.004	54.4 ± 1.7	61.8 ± 6.0%	24	3.1 ± 1.0	1998	1995	2000
	BON9	0.082 ± 0.009	84.4 ± 6.4	69.7 ± 8.5%	27	2.9 ± 0.7	1995	1992	1998
	BON10	0.099 ± 0.006	73.7 ± 2.9	57.5 ± 6.7%	31	2.9 ± 0.4	1991	1988	1995
<b>Average Bonete</b>	0.0924 ± 0.015			62.2 ± 12.2%					

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