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1 **^{210}Po sequential extraction applied to wetland soils at uranium mining sites**

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3

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10

11 **Abstract**

12 Former uranium mining activities have led to the presence of naturally occurring nuclides
13 embedded in soil. Such activities have also modified the secular equilibrium between
14 radionuclides in ^{238}U decay series. The objective of this paper is to quantify the long-term
15 effect of former uranium mining activities on the behavior of the final radionuclide in the
16 ^{238}U -series, i.e. polonium-210 (^{210}Po), present in soils. Soil samples are extracted from two
17 uranium sites in France, specifically a quarried site and a natural site. The polonium
18 distribution is studied within the various soil fractions, namely: water soluble, exchangeable,
19 bound to carbonates, bound to iron/manganese oxides, bound to organic matter, and residual.
20 ^{210}Po is mainly found in the residual fraction of both study sites (87-90%), followed by the
21 carbonates fraction (5-9%). The ^{210}Po activity in the other fractions is very small in
22 comparison with total activity.

23

24 **Keywords**

25 Polonium-210, Sequential extraction, Soils, Uranium mining.

26

27 1. Introduction

28 Over 200 uranium mines sites exist in France, some of which had been in operations until the
29 beginning of the 1990's (Ielsch *et al.*, 2017). Their existence has led to the presence of
30 naturally occurring nuclides embedded in soil. Given society's persistent concern about
31 radioactivity in the environment, assessing the impact of radioactive metals on environmental
32 compartments is necessary in order to adopt appropriate precautions. To quantify this impact
33 and understand the behavior of radionuclides in the environment, the main scientific challenge
34 calls for identifying the chemical mechanisms that control radionuclide mobility,
35 bioavailability and ecotoxicity. The major scientific milestone consists of quantifying labile
36 species in the ecosystem and then describing their speciation in terrestrial and aquatic
37 environments.

38 Discovered in 1898 by Pierre and Marie Curie, ^{210}Po is a natural radionuclide occurring in the
39 environment, with its precursors, i.e. radon-222 and lead-210, being the penultimate daughter
40 of natural uranium-238. Of the 33 polonium isotopes, ^{210}Po is one of the most abundant. Due
41 to its very high specific activity ($1.66 \cdot 10^{14} \text{ Bq.g}^{-1}$), ^{210}Po is a substance extremely toxic to
42 humans. Just a few micrograms are sufficient to cause death. This metallic substance is some
43 one million times more toxic than sodium or potassium cyanide (Al-Masri *et al.*, 2004;
44 Ansoberlo *et al.*, 2012). The issue of its bioavailability is directly correlated with the species'
45 chemical form, which depends on environmental characteristics (pH, Eh, organic carbon
46 content, etc.). This correlation between speciation and bioavailability remains puzzling given
47 the current state of knowledge on polonium behavior (notably as regards the limited
48 thermodynamic databases describing surface complexation reactions).

49 In the literature, many studies (Al-Masri *et al.*, 2006; Ekdal *et al.*, 2006; Karunakara *et al.*,
50 2000; Özden *et al.*, 2013; Strumińska-Parulska and Olszewski, 2018) demonstrate that
51 anthropic activities, such as phosphate industries, heavy metal mining and oil drilling, can
52 lead to a local 'overconcentration' of ^{210}Po . The correlation with uranium mining (i.e. ore
53 extraction) is most obvious, over the range of 0.014 to 22 kBq.kg⁻¹ on surface soil (Lapham
54 and Millard, 1989; Sethy *et al.*, 2014; Thomas, 2000). None of these authors could determine
55 whether the polonium present is labile. The fate of polonium in the environment will differ
56 because its behavior and mobility in the soil depend on its physicochemical forms as well as
57 soil characteristics (Blanco *et al.*, 2004; Jia *et al.*, 2006; Prakash *et al.*, 2017; Žemberyová *et*
58 *al.*, 2006).

59 A sequential extraction methodology allows access to the geochemical partitioning of heavy
60 metals or radionuclides within the various constituent solid phases of soils or sediments. This
61 methodology is based on a successive treatment of specific solutions, each designed to
62 solubilize a targeted mineralogical fraction (Vandenhove *et al.*, 2014). Despite problems such
63 as re-adsorption, non-selectivity of extraction reagents and an incomplete dissolution of target
64 phases (Schultz *et al.*, 1998), sequential extraction yields key information on the original
65 properties of radionuclides. ^{210}Po in mineral samples was speciated by applying sequential
66 extraction in order to identify their sources (Al-Masri *et al.*, 2006; Jia *et al.*, 2006; Štok and
67 Smodiš, 2012).

68 The aim of this study is to determine the distribution of ^{210}Po in the various geochemical
69 fractions of soil samples from two uranium sites in France, specifically an operational site
70 (Rophin) and a natural site (Granges du Colonel). For this purpose, a six-step sequential
71 extraction method has been applied. This methodology was validated using certified reference
72 materials from the IAEA Agency.

73

74 **2. Materials and methods**

75 *2.1. Study sites*

76 The two selected sites are in two enriched uranium zones in France (see Fig. 1). The first,
77 named ‘Rophin’, is located in the Puy-de-Dôme Department about 3.4 km northwest of the
78 city of Lachaux. The surrounding landscape is hilly and composed of forests and meadows.
79 The soil at the Rophin site is qualified as wet to marshy. Quarried from 1948 to 1957, the
80 Rophin site produced 0.4 tons of uranium for 2,000 tons of ore actually mined. This site now
81 serves as a storage depot for uranium waste and has not been remodeled since its inception in
82 the 1950's.

83 A mechanical or washing plant was built in 1948 on the mine itself in order to treat the
84 extracted material. The objective of this facility was to concentrate uranium ores from local
85 mining operations before processing.

86 The Rophin site is currently used to store ore processing residues and dismantling products
87 from laundries. The actual storage location lies on the former mining tile of the Rophin mine
88 and contains 30,000 tons of waste. Moreover, since this site has not been remodeled since its
89 initial construction in the 1950's, it is now naturally vegetated.

90 The second site, named ‘Granges du Colonel’, lies roughly 7 km northeast of Belvédère in the
91 Alpes-Maritimes Department. As part of the search for uranium resources in France, the
92 granitic massif in this department was prospected during the years 1956-58. The occurrences
93 of uranium here are mainly confined to the Permian lands, with the Granges du Colonel site
94 offering some of the highest indices. Yet this site has never been quarried, due perhaps to its
95 remote location and lack of accessibility or to local opposition. Samples from this site were
96 collected in order to compare the impact of human activities on the behavior of polonium in
97 the soil.

98

99 *2.2. Samples, sampling protocol, pretreatment*

100 **Certified material reference: IAEA-385**

101 The protocol validation and reproducibility steps were performed using the certified material
102 standard IAEA-385 with a given value of ^{210}Po . The material was collected in the Irish Sea by
103 the Centre for the Environment, Fisheries and Aquaculture Science (CEFAS), Lowestoft,
104 U.K. in 1995. It was then dried, sieved through a 250- μm mesh and homogenized by mixing

105 in a nitrogen atmosphere. This sample received International Atomic Energy Agency (IAEA)
106 certification (Pham *et al.*, 2005). The IAEA-385 sediment was subjected to the recognized
107 analysis procedure before application to our environmental samples.

108

109 **Gamma Cartography**

110 Gamma cartography was conducted at the study sites in order to identify areas of high
111 activity. Two gamma ray maps of the sites were derived from a gamma ray sensor with a
112 Colibri device (Canberra, Inc.). The data were acquired automatically every five seconds.
113 These measurements were recorded in May 2017 for the ‘Granges du Colonel’ site and in
114 November 2017 for ‘Rophin’. The gamma mapping images and photographs were
115 superimposed using the QGIS (Quantum Geographic Information System) software.

116

117 **Sampling and Sample Preparation**

118 Soil samples were collected in wetlands (denoted RW and GW for Rophin Wetland and
119 Granges du Colonel Wetland, respectively) with soil traps placed at a depth of 15 cm. After
120 sampling and storage in a plastic bag, the samples were dried in the oven at 50 °C for 10 days
121 followed by cooling at room temperature. Stones, pebbles, plant materials and roots were
122 manually removed. The samples were then sieved through a 2-mm screen sieve before being
123 crushed in a ceramic pestle and mortar to obtain a homogeneous mixture that was stored until
124 analysis.

125

126 *2.3. Analysis of soil properties*

127 **Soil pH**

128 The soil pH was measured by a method specified in NF ISO 10390 (AFNOR 1994b).
129 Measurements were carried out in two different media, i.e. distilled water or a 1 M KCl
130 solution (the cations in these solutions displace the protons absorbed on the soil ion
131 exchangers with as much quantitative precision as possible). Five grams of dry soil samples
132 were vigorously shaken with 25 ml of the corresponding solution for 5 minutes. The
133 suspension was then left to rest at least 4 hours before measurement with a pH meter
134 (SevenCompact™ METTLER TOLEDO).

135 **Carbonate and organic matter quantification**

136 The organic and carbonate matter contents in the soils were determined according to the LOI
137 (Loss On Ignition) method (Heiri *et al.*, 2001). Approximately 1 g of dried soil sample was
138 introduced into the oven at 50 °C overnight. Next, the soil samples were heated at 550 °C for
139 5 h in the muffle furnace (Nabertherm) to burn the organic matter. In a subsequent step, they
140 were heated to 950 °C for 2 h to burn the carbonate matter. Between each step, the samples
141 were cooled in a desiccator and weighed.

142 The organic content (%OM) was calculated using the following equation:

$$143 \quad \%OM = \frac{DW_i - DW_{550}}{DW_i} \times 100 \quad \text{Eq. 1}$$

144 For carbonate content (%CO₃²⁻), the calculation proceeded as follows:

$$145 \quad \%CO_3^{2-} = \frac{DW_{550} - DW_{950}}{DW_i} \times 100 \times 1.36 \quad \text{Eq. 2}$$

146 where DW_i denotes the dry weight of the sample before combustion, DW_{550} the dry weight of
147 the sample after heating at 550°C, and DW_{950} the dry weight of the sample after heating at
148 950°C (all in g). The weight loss at 950°C multiplied by 1.36 (ratio of carbonate to carbon
149 dioxide mass) equals the weight of the carbonate in the original sample.

150

151 2.4. ²¹⁰Po analysis

152 **Determination of total ²¹⁰Po activity in the soil**

153 0.5 g of soil with 40 mBq of an added ²⁰⁹Po (Eckert Ziegler) tracer was digested with 10 ml of
154 hydrofluoric acid (39%) in a microwave (Ethos Easy). The microwave cycle was set with two
155 steps: the first one raised the temperature to 200 °C for 20 minutes, while the second step
156 stabilized this temperature for 120 minutes. After cooling at room temperature, the
157 hydrofluoric acid was evaporated to dryness at 50 °C. Then, 10 ml of concentrated nitric acid
158 (65%) were added and then heated again with the same microwave cycle of mineralization.
159 Then, it is evaporated to dryness at 50 °C. For spontaneous polonium deposition onto a silver
160 disc, the residue was dissolved in a 0.5 M HCl solution before adding ascorbic acid (1 g for
161 10 ml HCl 0.5 M) (Nelson *et al.*, 2017; Özden *et al.*, 2013). The contact time was set for 24 h
162 at room temperature. The deposition was carried out in a nearly enclosed environment in
163 order to reduce the loss by evaporation (Jia *et al.*, 2006). The disc was then washed using
164 demineralized water and air-dried. Lastly, the polonium activity was measured by means of

165 alpha spectrometry. Triplicate samples were prepared to ensure the quality of the analysis
166 performed.

167

168 **Sequential Extraction Procedure**

169 The distribution of ^{210}Po in soil fractions was studied using a sequential extraction protocol
170 developed by Tessier *et al.* (Tessier *et al.*, 1979) and Al-Masri *et al.* (Al-Masri *et al.*, 2006).

171 This six-step procedure isolates the following fractions: water soluble, exchangeable, bound-
172 to-carbonates, bound-to-iron/manganese oxides, bound-to-organic matter, and residual
173 fraction. Triplicate samples were prepared in order to ensure analytical quality.

174 One gram of each soil sample was introduced into centrifuge cones (50 ml) and stirred at
175 room temperature for the first five steps. After each step, the suspension was centrifuged
176 (SigmaTM 10223) at 4,000 rev/min for 20 min. The supernatant was removed and filtered
177 through a 0.45- μm pore size cellulose filter. The extracted solutions were evaporated to
178 dryness at 50 °C. The residue in the centrifuge tube was then extracted by the reagent from
179 the next steps according to the following procedure.

180 *Fraction 1: Water soluble*

181 The samples were first extracted using 20 ml of demineralized water for 24 h in order to
182 release water-soluble ^{210}Po .

183 *Fraction 2: Exchangeable*

184 The residue from Fraction 1 was leached with 20 ml of 1 M NH_4OAc for 24 h. The ^{210}Po
185 adsorbed on mineral surfaces, capable of exchanging with an ion widely introduced into the
186 solution (NH_4^+), is released.

187 *Fraction 3: Bound-to-Carbonates*

188 The residue from Fraction 2 was contacted with a 20 ml buffered NaOAc/HOAc solution (pH
189 5.2 ± 0.1) for 6 h. At this pH, elements precipitated or co-precipitated with the carbonates, as
190 well as those that bind to the surfaces of solids by pH-dependent bonds, are released in
191 solution (Cornu and Clozel, 2000).

192 *Fraction 4: Bound-to-iron and manganese oxides*

193 The residue from Fraction 3 was contacted with 20 ml of 0.04 M NH₂OH.HCl for 24 h in
194 order to free ²¹⁰Po, which had been bound to iron and manganese oxides.

195

196

197 *Fraction 5: Bound-to-organic matter*

198 The residue from Fraction 4 was contacted with 15 ml of 30% H₂O₂ acidified to pH 2 by 0.02
199 M HNO₃ for 24 h; afterwards, 5 ml of 3.2 M NH₄OAc was added before separation so as to
200 avoid sorption. This extraction step destroys the organic materials in order to release the
201 complex or adsorbed metals.

202 *Fraction 6: Residual fraction*

203 Once the first five fractions had been removed, the residue consisted of the primary and
204 secondary minerals, i.e. the most difficult to extract (Rodgers *et al.*, 2015). The residue from
205 Fraction 5 was transferred to a Teflon tube and mineralized by the same procedure to
206 determine the total ²¹⁰Po activity concentration under the section 'Determination of total ²¹⁰Po
207 activity in the soil'.

208 ²⁰⁹Po was added to the filtrate resulting from each leaching step before evaporating to dryness
209 at 50 °C. The residue obtained was then dissolved using 0.5 M hydrochloric acid. The ²¹⁰Po
210 activity concentration in each fraction could be determined by alpha spectrometry after the
211 spontaneous deposition of polonium on the silver disc.

212 **Polonium quantification**

213 ²⁰⁹Po and ²¹⁰Po were quantified using dual alpha spectrometers EG&G Ortec 576A equipped
214 with boron-implanted silicon detectors offering a 450 mm² active area and an alpha resolution
215 (FWHM) of 20 keV at 5.47 keV. Pulses were analyzed with a multichannel buffer analyzer
216 (Spectrum Master Ortec 919) and spectra with the Vision software. The efficiency was
217 determined by means of calibrated discs electroplated with known amounts of tracer, while
218 the background noise was being controlled regularly. The average efficiency of the detector
219 was 22 ± 2%. The lower limit of detection for the method employed was 0.2 mBq (Leclercq
220 *et al.*, 2014).

221

222 3. Results and discussion

223 3.1. Total ²¹⁰Po activity in soil

224 The soil mineralization procedure using the microwave oven was performed with the IAEA-
225 385 sediment standard in order to validate this protocol. The recommended specific activity of
226 ²¹⁰Po for IAEA-385 was 28.0 Bq.kg⁻¹, with a confidence interval ranging from 23.8 to 36.6
227 Bq.kg⁻¹ dry weight. Table 1 shows the results of the total ²¹⁰Po activity concentration for the
228 standard IAEA-385 obtained from this work. The average activity found equals: 27 ± 3
229 Bq.kg⁻¹ dry weight. This value lies within the confidence interval reported by IAEA, thus
230 showing good agreement between the determined values and IAEA values (28.0 Bq.kg⁻¹). The
231 results of replicated measurements attest to the reproducibility of the procedure used.
232 Consequently, analyses of the standard IAEA-385 confirm that the recommended
233 mineralization procedure to determine total ²¹⁰Po amount yields reliable results.

234 Figure 2 presents the gamma ray map for both the Rophin (A) and Granges du Colonel (B)
235 sites. At Rophin, a stream flows down a gradient of the uranium mining zone and
236 subsequently merges with a north-south watercourse. The combined outflow of the two
237 currents is named the Terrasson stream. A radionuclide accumulation zone was identified
238 through this gamma mapping adjacent to the Terrasson stream 150 m after the confluence.
239 Gamma radiation activity in this area reaches 3500 c.s⁻¹, which is 7 times higher than the site
240 background level. The gamma irradiation levels at the Granges du Colonel site are much
241 lower than at Rophin.

242 A soil sample denoted RW (for Rophin Wetland) was extracted from a wetland where
243 radionuclide activity was locally at its highest level. A Granges du Colonel wetland soil
244 sample (GW) was also extracted for comparative purposes. Both samples were taken at depths
245 of 15 cm.

246 This mineralization protocol was then applied to the RW and GW samples. Table 1 reports
247 their respective ²¹⁰Po activity concentration (Bq.kg⁻¹ dry weight) levels. In the RW sample,
248 the ²¹⁰Po level was more than 40 times greater than that of GW, i.e. the ²¹⁰Po activity
249 concentrations in the GW and RW soil samples are 82 ± 4 (Bq.kg⁻¹) and $3,461 \pm 148$ (Bq.kg⁻¹),
250 respectively. These results are consistent with output from the gamma maps (Fig. 2). The
251 much higher ²¹⁰Po activity concentration of the Rophin sample can be explained by the
252 presence of a high radiation background emanating from the uranium storage facility at
253 Rophin.

254 3.2. *Sequential Extraction Studies*

255 According to Tessier *et al.* (Tessier *et al.*, 1979) and Al-Masri *et al.* (Al-Masri *et al.*, 2006), a
256 modified procedure was performed in order to detect the ^{210}Po distribution within the
257 geochemical fraction. The IAEA-385 standard was studied for ^{210}Po speciation using the
258 procedure of sequential extraction; results are shown in Table 2. It can be noticed that the
259 ^{210}Po fraction proportion of the sample equals: 1.24% in water-soluble form, 1.71% in
260 exchangeable form, 5.61% bound to carbonates, 2.92% bound to iron and manganese oxides,
261 1.09% bound to organic matter, and 87.4% bound to residue. In other words, ^{210}Po was almost
262 entirely found in the last fraction, thus indicating an association with the insoluble mineral
263 phases. The sum of ^{210}Po activity concentrations in these six each individual sequential
264 extraction fractions is equal to $25 \pm 3 \text{ Bq.kg}^{-1}$. This is in good agreement with the total ^{210}Po
265 activity measured in the unfractionated sample of $27 \pm 3 \text{ Bq.kg}^{-1}$. According to results
266 obtained from the standard sediment IAEA-385, this sequential extraction procedure has
267 successfully identified the ^{210}Po distribution in the various soil fractions.

268 This extraction procedure was then applied to GW and RW samples. The ^{210}Po activity
269 concentrations in the different soil fractions are presented in Table 2. Sequential extraction
270 results from the GW and RW samples are similar to those of the IAEA-385 sample. ^{210}Po was
271 mainly found in the residual fraction, followed by the carbonate fraction. The sum of ^{210}Po
272 activity in the four other fractions is relatively low, not exceeding 8% of total activity.

273 In a water-soluble and exchangeable fraction, the quantity of ^{210}Po was very small in
274 comparison with the other fractions. It can be concluded that polonium mostly attaches to
275 insoluble compounds in the atmosphere after the decay of radon gas (Al-Masri *et al.*, 2006).
276 The ^{210}Po was then deposited onto the ground as dry and wet deposition, which constitutes
277 their main source in surface soil (Karunakara *et al.*, 2000).

278 Apart from the residual fraction, ^{210}Po is mainly present in the carbonate fraction. However,
279 the percentage of ^{210}Po in the carbonate fraction of RW is much higher than that of GW, i.e.
280 88% and 43% respectively, by comparing it with the sum of the first five fractions. While the
281 carbonate content of both samples is equal (1.8% dry weight - Table 3). The uranium mining
282 activities promoted the exhalation of radon. This phenomenon has consequently modified the
283 secular equilibrium between radionuclides in the ^{238}U decay series. Radiochemical
284 disequilibrium points to the radionuclide mobilization and transport processes (Bister *et al.*,
285 2015). Elevated levels of ^{210}Po activity in the carbonates fraction can result from the decay of

286 unsupported radon-222; therefore, uranium mining activities at the Rophin site may indeed
287 influence the geochemical distribution of ^{210}Po . The increase in hydrogen ion concentration
288 changes the mobilization intensity of heavy metals. A low soil pH value, in addition to
289 mobilizing heavy metals, affects the activity of metal ions in the water contained in soil pores,
290 thus also altering their bioavailability (Fijałkowski *et al.*, 2012). The pH value of RW is less
291 than that of GW (Table 3), which may explain the higher portion of ^{210}Po found in the
292 carbonate-bound fraction in the RW sample.

293 ^{210}Po displays a low percentage (less than 1%) when $\text{NH}_2\text{OH}\cdot\text{HCl}$ was introduced during the
294 fourth step of the procedure, which would suggest that this element is only slightly associated
295 with iron and manganese oxides.

296 As regards the organic fraction, numerous studies (Avadhani *et al.*, 2005; Narayana *et al.*,
297 2006; Özden *et al.*, 2013) have shown a positive correlation between organic matter content
298 and ^{210}Po activity: the higher the organic matter content, the greater the level of ^{210}Po activity.
299 In contrast with these observations, results from the sequential extraction in this study reveal
300 that ^{210}Po activity concentration in the organic fraction remains very small, 0.16% and 3.43%
301 for RW and GW, respectively, whereas the organic matter content in RW and GW amounts to
302 respectively 22.7% and 9.2%. Prakash *et al.* found that the lower the organic matter
303 percentage in the soil samples, the higher the level of ^{210}Po activity and *vice versa* (Prakash *et*
304 *al.*, 2017). The organic matter and ^{210}Po were detected in the same location, i.e. on the soil
305 surface, yet they remain uncorrelated. According to Vaaramaa, ^{210}Po in the organic layer most
306 probably originated in large part from atmospheric depositions, and no systematic correlation
307 appeared between these two elements (Vaaramaa *et al.*, 2010). ^{210}Po can indeed stem from
308 two activity sources: supported ^{210}Po from decay of the remaining radon-222 in the soil
309 matrix, and unsupported ^{210}Po due to the deposition of radon decay products from the
310 atmosphere (Matthews *et al.*, 2007; Özden *et al.*, 2013; Persson, 2014). Upon reaching the
311 soil surface as fallout from the atmosphere, ^{210}Po is rapidly adsorbed by the surface soil,
312 which accounts for the principal source of ^{210}Po in surface soil (Avadhani *et al.*, 2005; Jia *et*
313 *al.*, 2006). On the other hand, the soil surface is also a place where organic substances are
314 primarily found. This initial layer of soil is mainly composed of decomposed plant debris and
315 humic substances. The surface soil layer is also known to be the zone in which most
316 biological activity occurs.

317 The highest ^{210}Po portions were dissolved in mineral acids, with observed values equaling
318 90% and 87% for the RW and GW samples, respectively. This finding reflects immobilization

319 of the particular radionuclide, which is due to the fact that ^{210}Po has been incorporated into
320 crystalline silicates. The low ^{210}Po availability leads to the conclusion that its presence in the
321 environment is not hazardous for ingestion pathways, but only for inhalation of the particulate
322 materials (Desideri *et al.*, 2008).

323

324 **4. Conclusion**

325 The total ^{210}Po activity concentration in soil samples extracted from two wetland areas near
326 French uranium mining sites has been determined. The sample taken near a uranium storage
327 facility reveals a significant level of total ^{210}Po activity; this finding is correlated with the high
328 radiation background. To study the distribution of ^{210}Po in soils, a sequential extraction
329 procedure was developed, validated with the certified reference material (IAEA-385) and then
330 applied to soil samples from the two study sites. From these sequential extraction analyses,
331 the ^{210}Po found was almost entirely in the residual fraction of both sites. Apart from the
332 residual fraction, ^{210}Po is mainly present in the carbonate-bound fraction; however, the
333 percentage of ^{210}Po in the RW sample exceeds that of GW even though the carbonate content
334 of both samples is equal. Elevated levels of ^{210}Po activity in the carbonate-bound fraction may
335 result from the decay of unsupported ^{222}Rn . Underground mining operations can thus provide
336 drains for the circulation of radon gas. This phenomenon modifies the secular equilibrium
337 between radionuclides in ^{238}U decay series.

338

339

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344

345

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442 **Figure and Table captions**

443 Fig. 1: Map of the mean uranium content of geological units in France from (Ielsch *et al.*,
444 2017), with identification of our two experimental sites

445

446 Fig. 2: Gamma cartography of the "Rophin" site (A) and the "Granges du Colonel" site (B)

447

448 Fig. 3: Distribution of ^{210}Po (%) in six fractions from the sequential extraction of IAEA-385,
449 RW and GW samples

450

451 Table 1: Total ^{210}Po activity concentration (Bq.kg^{-1} dry weight)

452

453 Table 2: Fractionation of ^{210}Po in the study samples ($\text{Bq.kg}^{-1} \pm \text{SD}$)

454

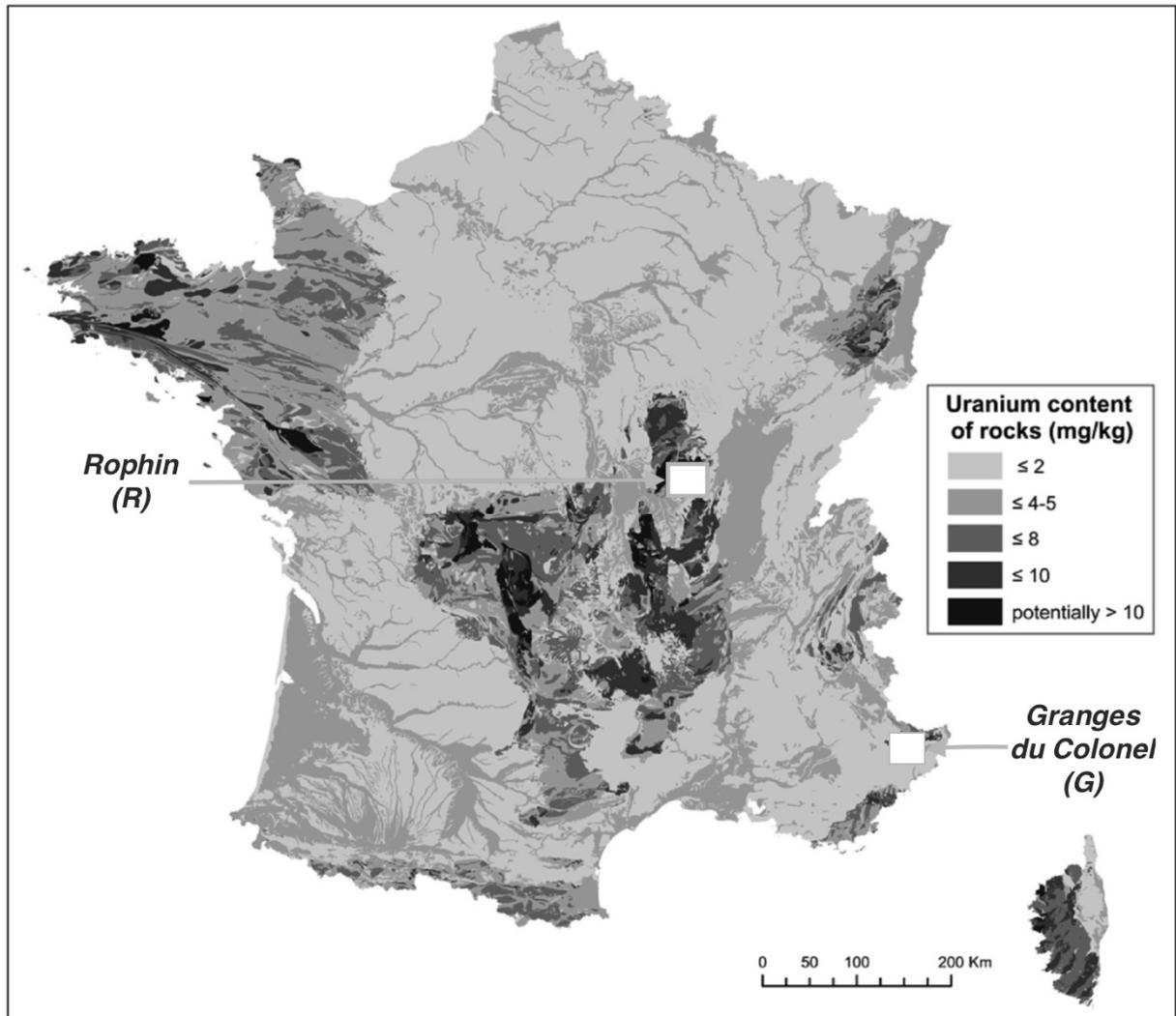
455 Table 3: Properties of the studied soil samples

456

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458 **List of figures and table**

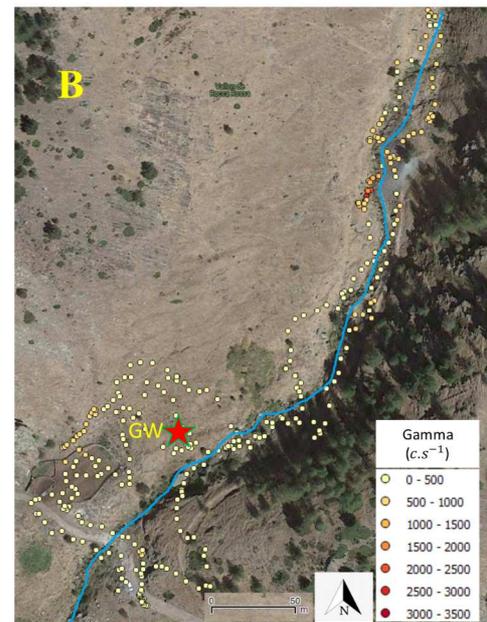
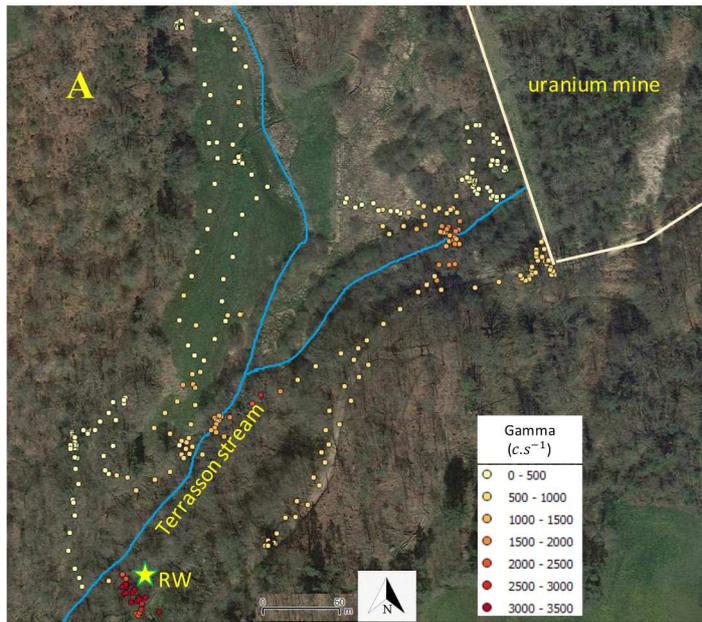
459 Fig. 1:



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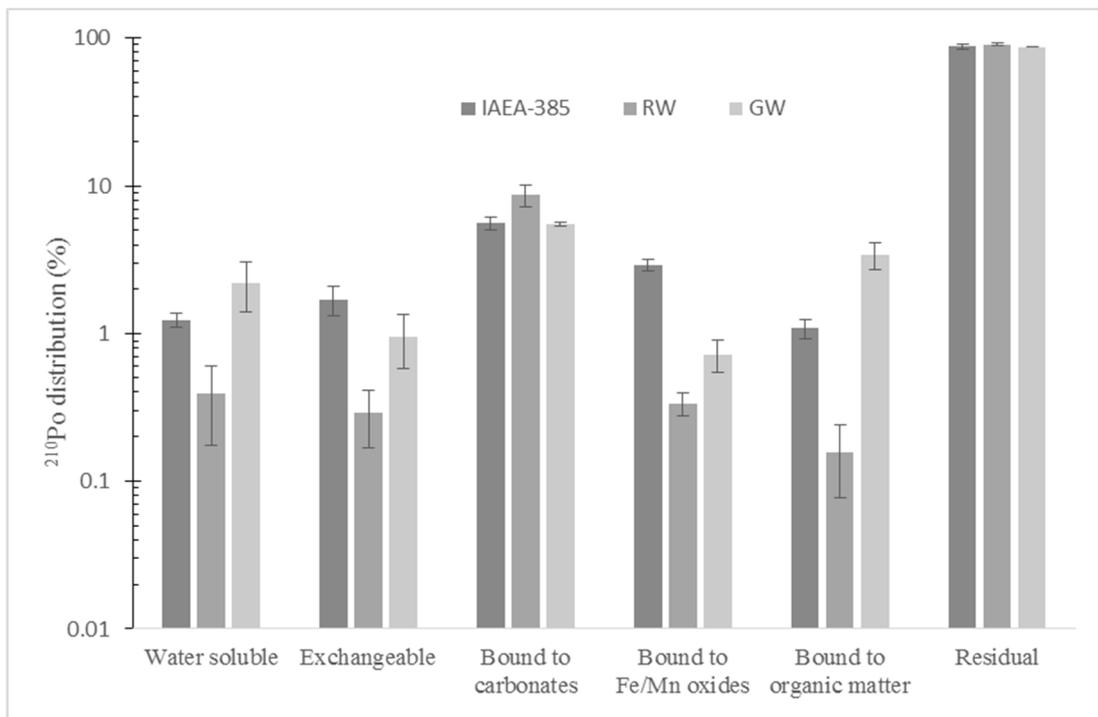
462 Fig. 2:



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465 Fig. 3:



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467

468 Table 1:

Sample	Activity concentration \pm SD
IAEA-385*	27 ± 3
RW	$3,461 \pm 148$
GW	82 ± 4

469 *Information value of ^{210}Po : 28.0 Bq.kg^{-1} with a confidence interval from 23.8 to 36.6 Bq.kg^{-1} dry weight

470

471 Table 2:

Fraction	IAEA-385	RW	GW
Water soluble	0.32 ± 0.03	12 ± 7	1.8 ± 0.6
Exchangeable	0.43 ± 0.09	9 ± 4	0.8 ± 0.3
Bound to carbonates	1.42 ± 0.2	266 ± 51	4.4 ± 0.3
Bound to Fe/Mn oxides	0.72 ± 0.06	10 ± 2	0.6 ± 0.1
Bound to organic matter	0.28 ± 0.06	5 ± 3	2.8 ± 0.7
Residual fraction	22 ± 2	$2,762 \pm 71$	70 ± 3

472

473

474 Table 3:

Sample	pH (H ₂ O)	pH (KCl)	%OM	%CO ₃ ²⁻
RW	5.51	3.96	22.7	1.8
GW	5.61	4.52	9.2	1.8

475