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Substratum influences uptake of radium-226 by plants

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20 **Abstract**

21

22 Radium-226, an alpha emitter with half-life 1600 years, is ubiquitous in natural environments.
23 Present in rocks and soils, it is also absorbed by vegetation. The efficiency of ^{226}Ra uptake by plants
24 from the soil is important to assess for the study of heavy metals uptake by plants, monitoring of
25 radioactive pollution, and the biogeochemical cycle of radium in the Critical Zone. Using a
26 thoroughly validated measurement method of effective ^{226}Ra concentration (EC_{Ra}) in the laboratory,
27 we compare EC_{Ra} values of the plant to that of the closest soil, and we infer the ^{226}Ra soil-to-plant
28 transfer ratio, R_{SP} , for a total of 108 plant samples collected in various locations in France. EC_{Ra}
29 values of plants range over five orders of magnitude with mean (min–max) of 1.66 ± 0.03 (0.020–
30 113) Bq kg^{-1} . Inferred R_{SP} values range over four orders of magnitude with mean (min–max) of
31 0.0188 ± 0.0004 (0.00069–0.37). The mean R_{SP} value of plants in granitic and metamorphic context
32 (0.073 ± 0.002 ; $n = 50$) is significantly higher (12 ± 1 times) than that of plants in calcareous and
33 sedimentary context (0.0058 ± 0.0002 ; $n = 58$). This difference, which cannot be attributed to a
34 systematic difference in emanation coefficient, is likely due to the competition between calcium and
35 radium. In a given substratum context, the compartments of a given plant species show coherent
36 and decreasing R_{SP} values in the following order (acropetal gradient): roots > bark > branches and
37 stems \approx leaves. Oak trees (*Quercus* genus) concentrate ^{226}Ra more than other trees and plants in this
38 set. While this study clearly demonstrates the influence of substratum on the ^{226}Ra uptake by plants
39 in non-contaminated areas, our measurement method appears as a promising practical tool to use for
40 (phyto)remediation and its monitoring in uranium- and radium-contaminated areas.

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46 **Keywords:** radon emanation; radium concentration; radium uptake; soil-to-plant transfer; pollution;

47 substratum

48

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49 **Highlights**

- 50 • Effective ^{226}Ra concentration (EC_{Ra}) from plants in non-contaminated areas
- 51 • EC_{Ra} of 108 plant samples and nearby local top soil samples measured
- 52 • Mean ^{226}Ra soil-to-plant transfer ratio (R_{SP}) is 0.0188 ± 0.0004 (about 2%)
- 53 • Significant substratum effect with 12 ± 1 higher R_{SP} on granite than on limestone
- 54 • Competition between calcium and radium thus revealed in non-contaminated areas

55 **1. Introduction**

56

57 The Critical Zone (CZ), which is the domain between the top of the aquifer and the top of
58 the canopy, is the part of the Earth's surface sustaining life. One fundamental issue currently is to
59 assess the fluxes of energy and matter between the various compartments of the CZ and their
60 vulnerability (Brantley et al., 2007; Lin, 2010). Soil, a key compartment at the boundary between
61 the lithosphere, biosphere, and atmosphere, sensitive to natural and unnatural forcing and essential
62 for mankind, often appears as the most polluted part of the CZ (Abrahams, 2002; Banwart, 2011).
63 Various pollutants, harmful to the biosphere, have been detected in soil, including heavy metals and
64 radionuclides (*e.g.*, He and Walling, 1996; Manta et al., 2002; Douay et al., 2009; Li et al., 2014;
65 Girault et al., 2016). Vegetation growing on top of this soil is directly capable of capturing naturally
66 occurring elements as well as pollutants in its roots, tissues, flowers and fruits (*e.g.*, Sheppard and
67 Evenden, 1988b; Carini, 1999; Ehlken and Kirchner, 2002). This uptake of tracer elements can also
68 be a way to constrain the biogeochemical cycle of elements from the pedosphere to the biosphere in
69 the CZ.

70 Ubiquitous in the environment, the alkaline earth element radium is present in all CZ
71 compartments including water, rock, soil, and vegetation. The radium-226 isotope belongs to the
72 uranium-238 decay chain and is an alpha emitter with a half-life of 1600 ± 7 years (Duchemin et al.,
73 1994). Several studies have focused on the uptake of ^{226}Ra by plants, quantifying a transfer factor
74 (or concentration ratio) of ^{226}Ra from the soil to the plant tissues (Simon and Ibrahim, 1990;
75 Sheppard et al., 2006; Vandenhove et al., 2009; Uchida et al., 2009). Predominantly, because of the
76 need to assess the radium content of agricultural products and the risk to the population, researches
77 on the ^{226}Ra uptake by plants have been carried out in ^{226}Ra -contaminated areas (*e.g.*, Simon and
78 Ibrahim, 1990 and references herein), such as former or operating uranium mining and milling sites
79 (Marple, 1980; Vasconcellos et al., 1987; Bettencourt et al., 1988; Ibrahim and Whicker, 1992;

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80 Markose et al., 1993; Madruga et al., 2001; Blanco Rodríguez et al., 2002, 2010 ; Vera Tomé et al.,
81 2002; 2003; Chen et al., 2005; Ryan et al., 2005; Soudek et al., 2007a,b, 2010; Carvalho et al.,
82 2009; Černe et al., 2011; Medley et al., 2013; Hu et al., 2014; Medley and Bollhöfer, 2016; Yan and
83 Luo, 2016), phosphate fertilizer processing complexes (Paul and Pillai, 1986; Martínez-Aguirre and
84 Periañez, 1998), radium salt factories (Bettencourt et al., 1988), depleted uranium ammunition sites
85 (Popovic et al., 2008), and other industrial units (Paul and Pillai, 1986). These studies have been
86 complemented by experiments on artificially ^{226}Ra -enhanced soils at the laboratory scale in pots or
87 at larger scale in lysimeter and field experiments (Gerzabek et al., 1998; Bunzl and
88 Trautmannsheimer, 1999; Vandenhove et al., 2005; Vandenhove and Van Hees, 2007; Nezami et
89 al., 2016), providing valuable insights for phytoremediation of contaminated areas (Thiry and Van
90 Hess, 2008; Vera Tomé et al., 2008, 2009; Abreu et al., 2014). By contrast, fewer studies have
91 focused on ^{226}Ra uptake by plants in non-contaminated areas or control sites (Sam and Eriksson,
92 1995; Ham et al., 2001; Karunakara et al., 2003; Pulhani et al., 2005; Popovic et al., 2008; da
93 Conceição et al., 2009; Lauria et al., 2009; Uchida and Tagami, 2009; Dragović et al., 2010; James
94 et al., 2011; Medley et al., 2013; Asaduzzaman et al., 2014; Al-Hamarneh et al., 2016; Mrdakovic
95 Popic et al., 2020). Despite these efforts, the uptake of ^{226}Ra by plants thus remains insufficiently
96 well understood.

97 The first mechanism of uptake of elements by plants is passive and results from element
98 concentration in groundwater and evapotranspiration. In this mechanism, ^{226}Ra accumulates in
99 leaves and can also be excreted (*e.g.*, Weis and Weis, 2004). In addition, uptake of elements by a
100 living plant is part of the metabolic cycle. In vascular plants, ^{226}Ra uptake takes place through the
101 various steps of the biological processes (*e.g.*, Simon and Ibrahim, 1990): mobility of Ra^{2+} ions
102 including release and diffusion from the solid phase to the soil solution, exchange of available Ra^{2+}
103 ions by sorption/desorption onto the surfaces of roots, transport of Ra^{2+} ions across membranes in
104 the roots, and diffusion and translocation of ^{226}Ra into plant tissues. Among other factors, it has

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105 been recognized that radium uptake depends on the presence of other alkaline earth elements of
106 smaller ionic radius such as barium, strontium, calcium, and magnesium. This suggests that the
107 radium uptake by plants decreases as the concentration of other alkaline earth elements in soil
108 increases, and that incorporation by roots can saturate (Nathwani and Phillips, 1979; Marple, 1980;
109 Simon and Ibrahim, 1987). Generally, a bottom-to-top decreasing gradient (*i.e.*, acropetal) of
110 radium concentration has been observed in plant tissues, from roots to stems and from stems to
111 shoots (*e.g.*, Simon and Ibrahim, 1990). As a testing hypothesis, we could consider that the
112 variability in ^{226}Ra uptake by plants may be due to different types of soil (substrate) and substratum,
113 in particular in non-contaminated areas. However, surprisingly, only small differences or no change
114 at all have been evidenced (*e.g.*, Simon and Ibrahim, 1987, 1990; Vera Tomé et al., 2003; Pulhani et
115 al., 2005).

116 Several methods, such as gamma-ray spectrometry, alpha-particle spectrometry, liquid
117 scintillation counting, and mass spectrometry, are commonly used to measure high ^{226}Ra levels of
118 numerous materials. However, for low ^{226}Ra levels in soil and for plant samples of relatively small
119 mass, such methods generally give large analytical uncertainty and data have remained limited.
120 Thus, to study ^{226}Ra uptake by plants in non-contaminated areas, an alternative technique is desired,
121 able to reach low ^{226}Ra levels for large amount of samples, in a cost-effective manner. A candidate
122 high-sensitivity technique with well-constrained leakage effects and relatively small uncertainty for
123 low ^{226}Ra levels is available; it is based on radon-222 emanation, as already suggested thirty years
124 ago (Simon and Ibrahim, 1990). Radon-222 is a radioactive gas (half-life 3.8 days) produced by the
125 alpha decay of ^{226}Ra . The probability that a ^{226}Ra atom decays into a ^{222}Rn atom able to escape from
126 a medium is the emanation coefficient E (Tanner, 1964; Nazaroff, 1992). We define the ^{222}Rn
127 emanating power of a given material by the effective ^{226}Ra concentration (EC_{Ra}), *i.e.* the product of
128 E by the bulk ^{226}Ra concentration (C_{Ra}), expressed in Bq kg^{-1} (Stoulos et al., 2004). Based on the
129 accumulation method, EC_{Ra} has been measured in various materials including soils (*e.g.*,

130 Markkanen and Arvela, 1992; Girault et al., 2011; Perrier et al., 2016b), rocks and building
131 materials (e.g., Przylibski, 2000; Righi and Bruzzi, 2006; Hassan et al., 2011; Girault et al., 2012),
132 and more recently plants (Perrier et al., 2018). Lately, this method has been updated with a
133 significantly higher sensitivity, allowing EC_{Ra} measurement of material with small mass (< 5 g) and
134 low ^{226}Ra levels ($< 10^{-14}$ g g $^{-1}$) (Girault et al., 2017a; Girault and Perrier, 2019). Measuring EC_{Ra} of
135 plants using this high-sensitivity method is particularly suited in areas characterized by low ^{226}Ra
136 levels.

137 In this paper, to test the hypothesis of a possible effect of the substratum on the ^{226}Ra uptake
138 by plants in non-contaminated areas, we present results of effective ^{226}Ra concentration (EC_{Ra}) in a
139 total of 108 plants collected at several non-contaminated sites in France that belong to two
140 geological subsets: granitic and metamorphic context, and calcareous and sedimentary context.
141 Using measured EC_{Ra} of plants and of the nearby soil as well as representative ^{222}Rn emanation
142 coefficients for plants and soils, we infer the ^{226}Ra soil-to-plant transfer ratio (R_{SP}). We show that,
143 by contrast with the results available previously in contaminated areas, R_{SP} values strongly depend
144 on the substratum. We then discuss our results in terms of plant type, species, and compartment, and
145 of the ^{226}Ra concentration of soil–plant pairs. Consequences for the assessment of element fluxes in
146 the CZ are discussed in the conclusion.

147

148 **2. Material and method**

149

150 *2.1. Plant and soil samples*

151

152 A total of 108 plant samples and their associated nearby local soil samples were collected at
153 different sites in France (Fig. 1). The plants mainly include deciduous trees (*Quercus robur*, *Fagus*
154 *sylvatica*, *Castanea sativa*, *Tilia xeuropaea*, *Prunus cerasus*, *Ficus carica*, *Fraxinus excelsior*,

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155 *Corylus avellana*, *Aesculus hippocastanum*), evergreen trees (*Quercus ilex*, *Pinus pinaster*, *Abies*
156 *alba*), shrubs (*Buxus sempervirens*, *Phillyrea latifolia*, *Spartium junceum*, *Cytisus*
157 *oromediterraneus*), and ferns (*Pteridium aquilinum*). Other samples, such as market vegetables (n =
158 15), mosses (n = 5), mushrooms (n = 4), and algae (n = 1), were also collected.

159 This sample set is divided into two subsets based on geology: the A subset corresponding to
160 plants growing in granitic and metamorphic context (Fig. 1; in red), and the B subset corresponding
161 to plants growing in calcareous and sedimentary context (Fig. 1; in blue). Samples from the A
162 subset (granitic and metamorphic context; n = 50) come from: the calco-alkaline biotite Hercynian
163 granite of the Mont-Lozère mountain in the Sapine watershed, Lozère department (44.356567°,
164 3.808383°, 1174 m); the Cévennes granite, a southward extension of the Mont-Lozère Pluton, near
165 Anduze, Gard department (44.077317°, 3.972301°, 201 m); the Hercynian Limousin (Massif
166 Central) Saint-Sylvestre granite near Ambazac, Haute-Vienne department (45.976100°, 1.392017°,
167 426 m); and the leptynite (laminated orthogneiss) of the Saint-Yrieix arc near Saint-Yrieix-la-
168 Perche, Haute-Vienne department (45.474583°, 1.138317°, 304 m). Samples from the B subset
169 (calcareous and sedimentary context; n = 58) come from: the Kimmeridgian limestone near
170 Puéchabon, Hérault department (43.738395°, 3.591345°, 271 m); the Callovo-Oxfordian limestone
171 of the Pech Merle cave near Cabrerets, Lot department (44.507217°, 1.643633°, 292 m); the
172 Triassic gypsum mixed with rocky slope debris from the Oxfordian limestone near Anduze, Gard
173 department (44.058753°, 3.979291°, 159 m); the Stampian Fontainebleau sand in the Paris Basin
174 near Vaugrigneuse, Essonne department (48.609549°, 2.104894°, 112 m); and the Burdigalian sand
175 and marls of the Orléans formation near Chambon-la-Forêt, Loiret department (48.024976°,
176 2.261318°, 152 m).

177 At the selected sites, the radium concentration in groundwater can be considered negligible
178 and the main source of radium is the soil or the regolith. This hypothesis was confirmed at the sites
179 where ^{226}Ra concentration in groundwater could be measured (Perrier et al., 2016a): 4.8 ± 0.4 mBq

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180 L⁻¹ (n = 4) in the Sapine watershed, 4.2 ± 0.3 mBq L⁻¹ (n = 9) in Anduze, < 1 mBq L⁻¹ (n = 1) in
181 Pech Merle, 2.7 ± 0.6 mBq L⁻¹ (n = 3) in Vaugrigneuse, and 15.4 ± 1.6 mBq L⁻¹ (n = 2) in
182 Chambon-la-Forêt. These values belong to the lower range of ²²⁶Ra concentration in groundwater
183 (Girault et al., 2018).

184 Plant samples were directly collected in the field using clean scissors or shears and placed in
185 plastic bags. For some plants, several samples were collected from different compartments. Then,
186 plant samples, without washing with water, were gently dried in the laboratory at ambient room
187 temperature and cut in centimeter-size pieces. Top soil samples of mass 100–150 g were collected
188 nearby each plant sample in a systematic manner (plant–soil matched pairs) using a clean shovel
189 and placed in plastic bags. At some sites, several plant samples can be associated with one
190 representative soil sample. Only large pieces of stones were removed from soil samples at the time
191 of sampling, and no sieving, milling or homogenization were performed afterward in the laboratory.
192 Soil samples were also dried in the laboratory at ambient room temperature, as oven drying can
193 affect the value of EC_{Ra} (Girault and Perrier, 2011, 2012a), in order to keep them as much as
194 possible in the original natural condition.

195

196 *2.2. Measurement of water pH of soil samples*

197

198 Soil pH was determined in the laboratory using the common technique (*e.g.*, Thu et al.,
199 2020). About 20 g of soil and 100 mL of distilled water were placed in a beaker and stirred during
200 20 to 30 min with a magnetic stirrer. The pH of the suspension solution obtained was measured at
201 about 25°C using a regularly calibrated pH211 Microprocessor pH-meter (Hanna Instruments,
202 USA) an hour minimum after stirring to ensure equilibrium. The pH value of a total of 18

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203 representative soils of the two subsets was measured. Absolute experimental uncertainties
204 associated with calibration and reproducibility ranged from 0.01 to 0.02.

205

206 2.3. Measurement of effective ^{226}Ra concentration (EC_{Ra})

207

208 A mass m of gently dried plant or soil sample was placed in a hermetically closed container,
209 *i.e.*, a glass pot or bottle closed with a natural rubber stopper (Girault and Perrier, 2012a; Perrier et
210 al., 2018). For all samples, after an accumulation time, t , from 4 to 18 days, the air of the pot was
211 sampled using a 125-mL pre-evacuated scintillation flask (Algade, France). After the 3.5 h needed
212 to reach radioactive equilibrium, a photomultiplier (CALENTM, Algade, France) was used to
213 measure the number of counts in 10 min interval. Subtracting the background count of the flask
214 determined before sampling, applying the conversion factor of the photomultiplier, and taking into
215 account dilution during sampling (Girault and Perrier, 2012a), we infer ^{222}Rn activity concentration
216 in the flask, C_{Rn} (in Bq m^{-3}), and then the effective ^{226}Ra concentration (EC_{Ra}) of the material in the
217 container using (Girault and Perrier, 2011, 2012a):

218

$$EC_{\text{Ra}} = \frac{V}{m} \frac{C_{\text{Rn}}}{(1 - e^{-\lambda t})}, \quad (1)$$

219 where V is the free air volume of the container (m^3) and λ is the ^{222}Rn decay constant ($2.1 \times 10^{-6} \text{ s}^{-1}$).
220 Three accumulation experiments giving three EC_{Ra} measurements were performed for each sample
221 at different accumulation times and their values were averaged to get the final value. The
222 measurement uncertainty takes into account the counting statistics and the dilution correction. A
223 systematic uncertainty of about 5%, common to all measurements, is due to the absolute calibration
224 of the flask counting in the photomultipliers.

225 When ^{226}Ra level was particularly low, a high-sensitivity method was used (Girault et al.,
226 2017a; Girault and Perrier, 2019). In this method, the same protocol was followed, except that air

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227 sampling was conducted after a long accumulation time of more than 21 days in order to reach
228 radioactive equilibrium between ^{222}Rn and ^{226}Ra . The number of counts in the flask was then
229 recorded in 10 min interval in long counting session of 24 h. The background count was determined
230 precisely before sampling during a counting session of 3 days minimum. The final signal was
231 calculated by subtracting from the arithmetic average of the flask count distribution the arithmetic
232 average of the background distribution. Similarly, the effective ^{226}Ra concentration (EC_{Ra}) of the
233 material in the container was determined using Eq. (1). Generally, one or two additional high-
234 sensitivity measurements were performed for samples having an experimental uncertainty larger
235 than about 25% using the first method. For each sample for which significant EC_{Ra} values were
236 obtained using the two methods, or for which EC_{Ra} values were obtained using different subsamples
237 of the same sample, the final retained EC_{Ra} value is a weighted mean accounting for all the
238 available measurements.

239 In the last 10 years, more than 7300 measurements of EC_{Ra} have been performed in our
240 laboratory on various materials, based on more than 17,000 accumulation experiments, and we have
241 thoroughly tested the overall quality of our EC_{Ra} methodology. Based on the regularly measurement
242 of selected reference samples (Girault and Perrier, 2012a,b; Perrier et al., 2018), and on several
243 successful inter-comparison exercises with other measurement techniques from various laboratories
244 worldwide, such as three types of solid-state nuclear track detectors (Kodalpa™ LR115 cellulose
245 nitrate films from Dosirad, France; DPR2™ dosimeters from Algade, France; CR-39 polycarbonate
246 films from Kingston Univ., UK) (Girault and Perrier, 2012a,b), ionization chambers coupled with
247 alpha spectroscopy (AlphaGUARD™, Bertin Instr., Germany) (Nicolas et al., 2014; Girault et al.,
248 2017b), liquid scintillation counting (Wrocław Univ. of Technology, Poland) and alpha
249 spectrometry methods (CEA, France) (Perrier et al., 2016a), our EC_{Ra} measurement technique can
250 be considered as sufficiently robust and accurate for the present purpose, with the experimental
251 uncertainties assigned carefully.

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252 Using the two variants of our method (see above), we carried out a total of 225 EC_{Ra}
253 measurements of plant samples (Fig. 2), with mean (min–max) sample mass of 74 ± 4 (4.2–340) g
254 (Supplementary Fig. S1). The mean relative experimental uncertainty on plant EC_{Ra} was 21 ± 1 %
255 ($n = 225$), on average slightly larger than for all our other measurements that include mainly soil
256 and rock samples (11.0 ± 0.2 %; $n = 6636$). The mean relative experimental uncertainty on their
257 nearby soil EC_{Ra} was 2.85 ± 0.01 % ($n = 32$). For a plant sample mass between 15 and 50 g, the
258 relative experimental uncertainty was 4% for EC_{Ra} of 20 Bq kg^{-1} and about 30% for EC_{Ra} of 0.5 Bq
259 kg^{-1} (Fig. 2). Such measurement uncertainties are adequate for the present purpose.

260 The effects that can potentially affect, in principle, the interpretation of accumulation
261 experiments with rocks and soils (Girault et al., 2012b) could also be present for plant samples.
262 Temperature and humidity content have a significant effect on EC_{Ra} (e.g., Girault and Perrier, 2011,
263 2012b), but should be a second order effect in the temperature range that can be considered here for
264 plants. Physical surface adsorption of radon is largely unknown in the case of plant material, but
265 doping experiments suggested that it must be a second order effect (Perrier et al., 2018). Grain-size
266 may in principle affect EC_{Ra} values for solid materials (e.g., Markkanen and Arvela, 1992), but the
267 effect, if any, is unknown for plant. Considering beech leaves from the Sapine site (*Fagus sylvatica*;
268 A subset), EC_{Ra} values obtained for entire leaves ($8.19 \pm 0.50 \text{ Bq kg}^{-1}$) and for leaves cut in small
269 (<2 mm size) pieces ($8.44 \pm 0.57 \text{ Bq kg}^{-1}$) were compatible within uncertainty. Consequently, we
270 can consider here that the apparent EC_{Ra} that we measure in our experiments is representative of the
271 true EC_{Ra} .

272

273 2.4. Determination of ^{226}Ra soil-to-plant transfer ratio (R_{SP})

274

275 First, we compared the measured EC_{Ra} values of the plant sample (EC_{RaP}) and of the nearby
276 local soil (EC_{RaS}). We considered the plant–soil pair only when the nearby soil was not more distant

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277 than 100 m from the collected plant. On average, the distance between the plant and soil samples
278 was 6 ± 1 m. More than 80% of our plant samples have a soil sample within 10 m. Dispersion of
279 EC_{Ra} values for soil samples have been studied at several locations at various spatial scales and
280 were found reasonably compatible with the mean EC_{Ra} value of the given data set (Girault et al.,
281 2012b; Perrier et al., 2018). Some depth profiles (Supplementary Fig. S2), carried out in the soil
282 underlying the Mont-Lozère calco-alkaline biotite Hercynian granite in the Sapine watershed (A
283 subset) and in the soil underlying the Callovo-Oxfordian limestone of the Pech Merle cave (B
284 subset), show that EC_{Ra} values are relatively similar in the depth range 5 cm to 30 cm at a given
285 site. Two horizontal profiles at the Sapine site (A subset) do not show significant difference in EC_{Ra}
286 value of soil at distance of at least 60 m (Supplementary Fig. S3a). In addition, a 600-m-long
287 horizontal profile of EC_{Ra} values of soil carried out at Vaugrigneuse site (B subset), where 724 top
288 soils were sampled and their EC_{Ra} value measured (Perrier et al., 2016b), does not show any
289 significant variation of EC_{Ra} around the location of the soil where plants have been collected
290 (Supplementary Fig. S3b). These observations confirm that, at a given site, whatever the considered
291 subset, EC_{Ra} values of soil are relatively homogeneous, and the relation with plant sample in the
292 reasonable ranges of depth and lateral distance, as considered in our sampling methodology, can be
293 considered as representative of the mean EC_{Ra} values of the soil surrounding a given plant.

294 The obtained ratio (EC_{RaP}/EC_{RaS}) might be an interesting transfer parameter in itself. Indeed,
295 it is meaningful to normalize the effective ^{226}Ra concentration of the plant sample, which is to a first
296 approximation the ^{226}Ra concentration of the plant sample, as we will show below, to the effective
297 ^{226}Ra concentration of the soil, which represents the amount of ^{226}Ra connected to the pore space,
298 thus available to the root system. Nevertheless, in a second approach and in order to obtain values
299 of the soil-to-plant transfer ratio (R_{SP}), similar to the transfer factor or the concentration ratio
300 commonly used in the literature, we eliminate the mean emanation coefficients from the observed
301 ^{226}Ra plant-to-soil effective concentration ratios. Indeed, because EC_{Ra} gives an effective ^{226}Ra

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302 concentration of a given material, dividing EC_{Ra} of a material by its emanation coefficient, E , gives
303 the bulk ^{226}Ra concentration of the material. Here, we used mean representative E values for soils
304 and plants, as inferred from ^{226}Ra doping experiments carried out in the laboratory (Perrier et al.,
305 2018): $\langle E_S \rangle = 0.242 \pm 0.035$ for soils, and $\langle E_P \rangle = 0.862 \pm 0.044$ for plants. The R_{SP} value is then
306 calculated using (Perrier et al., 2018):

$$307 \quad R_{SP} = \frac{E_S}{E_P} \frac{EC_{RaP}}{EC_{RaS}}. \quad (2)$$

308 A total of 108 plant samples with a nearby local soil were collected and their respective R_{SP} value
309 could be inferred. Because it accounts for EC_{Ra} and E experimental uncertainties of both plant and
310 soil samples, uncertainty on R_{SP} was generally large, with mean value of $23 \pm 9 \%$. In the following,
311 averages are geometric means except otherwise stated.

312 In plant material, trapping of radon cannot occur as in minerals (Nazaroff, 1992; Adler and
313 Perrier, 2009), therefore the emanation coefficient of plants must be close to 1 and cannot vary
314 much from plant to plant. In the case of soils, however, the potential range of variations can be large
315 (Sakoda et al., 2011), and we shall return to the matter in details in the discussion below.

316

317 **3. Results**

318

319 *3.1. Effective ^{226}Ra concentrations in plants and their nearby local soils*

320

321 Effective ^{226}Ra concentration (EC_{Ra}) values of plant samples ($n = 108$) range over about five
322 orders of magnitude (Fig. 3c), from 0.020 ± 0.001 to $113 \pm 7 \text{ Bq kg}^{-1}$, and with a mean of $1.66 \pm$
323 0.03 Bq kg^{-1} (Table 1). The two largest values, 113 ± 7 and $44 \pm 2 \text{ Bq kg}^{-1}$, are measured for a moss
324 and roots of *Fagus sylvatica*, respectively, both from the granitic area near Ambazac. The two
325 smallest values, 0.020 ± 0.001 and $0.09 \pm 0.07 \text{ Bq kg}^{-1}$, are measured for chestnuts of *Aesculus*

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326 *hippocastanum* and branches of *Prunus cerasus*, respectively, both from the sedimentary area near
327 Anduze. The EC_{Ra} values of the nearby soil samples ($n = 24$) are less scattered (Fig. 3b) with a
328 mean of 18.9 ± 0.2 Bq kg⁻¹ (Table 1). While the EC_{Ra} values of plants follow a log-normal
329 distribution with two modes around 0.5 and 15 Bq kg⁻¹, similar to our whole data set of plant EC_{Ra}
330 ($n = 174$) (Fig. 3c), the EC_{Ra} values of soils follow a log-normal distribution with a single mode
331 around the mean (Fig. 3b). Although, to first order, we may expect some relation between EC_{Ra} of
332 plant and EC_{Ra} of soil, the mean value and the general shape of the log-normal distribution of plant
333 EC_{Ra} appear relatively similar to those of rock EC_{Ra} (Fig. 3a), suggesting a possible relation
334 between a given plant and its substratum. Alternatively, the large range of plant EC_{Ra} may also
335 suggest an efficient dispersion process during the transfer of ²²⁶Ra from soil to the biosphere.

336 The comparison of plant EC_{Ra} for the two subsets provides an important hint. Indeed, a
337 significant difference is observed between EC_{Ra} values of plants collected in granitic and
338 metamorphic context (A subset; mean: 9.2 ± 0.1 Bq kg⁻¹; $n = 50$) and EC_{Ra} values of plants
339 collected in calcareous and sedimentary context (B subset; mean: 0.38 ± 0.01 Bq kg⁻¹; $n = 58$).
340 Separating these two subsets, only few plant samples have similar or even slightly larger EC_{Ra} than
341 their respective soil EC_{Ra} (Fig. 4a). However, more plant samples have EC_{Ra} relatively similar to
342 their respective rock EC_{Ra} (Supplementary Fig. S4). Soil EC_{Ra} values are also different, depending
343 on the substratum: 36.1 ± 0.6 Bq kg⁻¹ for the A subset and 12.9 ± 0.2 Bq kg⁻¹ for the B subset (Table
344 1 and Fig. 4a). The EC_{Ra} of the A subset over EC_{Ra} of the B subset ratio is significantly larger for
345 plants (24 ± 8) and rocks (31 ± 1) than for soils (2.8 ± 0.1).

346

347 3.2. Results on the corrected ²²⁶Ra soil-to-plant transfer ratio

348

349 Normalizing plant EC_{Ra} by emanation coefficient of plants (E_P) and similarly soil EC_{Ra} by
350 E_S , we observe a slight EC_{RaP}/E_P increase with EC_{RaS}/E_S , from the B subset to the A subset (Fig.

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351 4b). The min–max values of EC_{RaP}/E_P and EC_{RaS}/E_S respectively, corresponding to the bulk ^{226}Ra
352 concentrations, are 0.6–130 Bq kg^{-1} and 50–370 Bq kg^{-1} for the A subset, and 0.02–5 Bq kg^{-1} and
353 11–190 Bq kg^{-1} for the B subset, which are consistent with reported ^{226}Ra concentration in plants
354 and soils in similar contexts.

355 The inferred values of soil-to-plant transfer ratio (R_{SP}) ($n = 108$) range over four orders of
356 magnitude (Fig. 5a), from 0.0007 ± 0.0001 to 0.37 ± 0.06 , and with a mean of 0.0188 ± 0.0004
357 (Table 1). Our R_{SP} values follow a log-normal distribution with two modes around 0.003 and 0.1
358 (Fig. 5a). Our whole R_{SP} data set can be subdivided into our two substratum subsets (Table 1).
359 Indeed, the highest mode of the R_{SP} distribution corresponds to the A subset (Fig. 5b), with R_{SP}
360 values from 0.005 ± 0.001 to 0.37 ± 0.06 and mean of 0.073 ± 0.002 ($n = 50$), while the smallest
361 mode of the R_{SP} distribution corresponds to the B subset (Fig. 5c), with R_{SP} values from $0.0007 \pm$
362 0.0001 to 0.13 ± 0.02 and mean of 0.0058 ± 0.0002 ($n = 58$).

363 While the discrimination between the two subsets appears significant, some tail-ends of the
364 R_{SP} distribution nevertheless exist for both subsets. About 20% of the A subset has a R_{SP} value
365 smaller than 0.02, while about 19% of the B subset has a R_{SP} value higher than 0.02. A better
366 discrimination, however, can be proposed for R_{SP} as a function of the inferred plant or soil ^{226}Ra
367 concentration (Fig. 4c and Supplementary Fig. S5). In Fig. 4c, defining the line by
368 $R_{SP}=1.24 \times 10^5 C_{RaS}^{-3.43}$, then 6% of the A sample now falls in the B region, while 5% of the B
369 samples falls in the A region. This suggests that the two subsets can be separated almost
370 completely. Overall, our mean R_{SP} value in granitic and metamorphic context is 12 ± 1 times larger
371 than that in calcareous and sedimentary context. This observation indicates definitely, for the first
372 time, a clear quantifiable effect of the substratum on the ^{226}Ra transfer from soil to plant in non-
373 contaminated areas.

374

375 4. Discussion

376

377 *4.1. Updated status on the understanding of radium uptake by plants*

378 Our soil-to-plant transfer ratio (R_{SP}) data, although they are inferred from the measurement
379 of effective ^{226}Ra concentration (EC_{Ra}) and emanation coefficient (E) of both soil and plant
380 samples, give compatible values with the transfer factors deduced from the direct measurement of
381 bulk ^{226}Ra concentrations as reported in the literature. Indeed, our mean R_{SP} value, 0.0188 ± 0.0004 ,
382 thus about 2%, is consistent with mean ^{226}Ra transfer factors reported in the literature for all plants,
383 ranging from about 2% (Vandenhove et al., 2009) to about 5% (Sheppard et al., 2006). Using a
384 compiled data set from published ^{226}Ra transfer factors from the literature (38 articles; $n = 870$),
385 90% of the R_{SP} values are comprised between 0.0015 and 1. R_{SP} values above 1 are generally
386 obtained with vegetables growing in radium-contaminated environments or in laboratory
387 experiments using radium-rich nutrient solutions (Simon and Ibrahim, 1990 and references herein;
388 Sam and Eriksson, 1995; Martínez-Aguirre and Perriñez, 1998; Vanderhove and Van Hees, 2007;
389 Asaduzzaman et al., 2014; Hu et al., 2014). R_{SP} values below 0.0015 are generally obtained with
390 vegetables, fruits, and cereals growing in radium-contaminated environments or, in a lesser extent,
391 in non-contaminated areas (Simon and Ibrahim, 1990 and references herein; Markose et al., 1993;
392 Ham et al., 2001; Ryan et al., 2005; Uchida and Tagami, 2007; Uchida et al., 2009 and references
393 herein; Černe et al., 2011). Considering a reasonable range of R_{SP} values over five orders of
394 magnitude (from 0.00025 to 2; $n = 844$), these R_{SP} values follow a log-normal distribution with one
395 mode, centered on the mean of 0.0448 ± 0.0003 (Fig. 5d). In the literature, only few R_{SP} values
396 smaller than 0.006 (about 11%) are reported. This small range corresponds to the low R_{SP} values we
397 obtained in calcareous and sedimentary context (B subset). To our knowledge, no study has focused
398 on radium transfer from soil to plants in calcareous and sedimentary context, probably because of
399 the low ^{226}Ra levels that are beyond the detection limit of the commonly used methods (gamma and
400 alpha spectrometry, liquid scintillation counting, and mass spectrometry). This highlights the

401 usefulness of our high-sensitivity EC_{Ra} measurement method, decreasing the detection limit of ^{226}Ra
402 levels in soil and plant samples with reasonable experimental uncertainty.

403 Our data and the data from the literature suggest that the transfer of ^{226}Ra from soil to plant
404 may be log-normal (Fig. 5). While the log-normal distribution of radioactivity data is a subject in
405 itself (Bossew, 2010), it could be that transfer of radionuclides and other trace elements to plants is
406 intrinsically affected by large fluctuations resulting in a log-normal distribution. The standard
407 deviation then is as interesting as the mean value and is an independent parameter. However, the
408 process controlling the standard deviation is not clear at this stage and a larger data set would be
409 required to evaluate the distribution function.

410 Contrary to other studies (*e.g.*, Simon and Ibrahim, 1987, 1990; Vera Tomé et al., 2003;
411 Pulhani et al., 2005), we have found a significant difference in R_{SP} depending on the substratum.
412 Our difference of a factor of 12 ± 1 between R_{SP} obtained in granitic and metamorphic context (A
413 subset) and R_{SP} obtained in calcareous and sedimentary context (B subset) may be explained by the
414 larger calcium concentration in the soil above limestone regolith. In the case of the Sapine
415 watershed (A subset), calcium deficit was reported in the soil (Durant et al., 1991; Didon-Lescot,
416 1996). In contrast, a large calcium concentration may saturate incorporation into the plant roots,
417 hence decreasing the transfer of ^{226}Ra from soil to plant, as reported in earlier studies (Nathwani
418 and Phillips, 1979; Marple, 1980; Simon and Ibrahim, 1990). Here, we reveal this effect for the first
419 time in non-contaminated natural environments. This observation suggests that other effects related
420 to substratum may influence R_{SP} of ^{226}Ra and other radionuclides or heavy metals as well.

421

422 *4.2. Radium uptake in the different plant compartments and in other biological samples*

423

424 For some plant species, we collected several plant compartments. When considering
425 separately the two subsets of samples in different substratum contexts, the R_{SP} values obtained for

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426 the different compartments of a given plant appear relatively compatible (Fig. 6 and Tables 2 and
427 3). The acropetal gradient, *i.e.*, a decrease of ^{226}Ra concentration from roots to apex, is generally
428 observed for all plants: the root-to-shoot ratio ranges from 0.7 ± 0.3 to 2.3 ± 1.2 , with a mean of 1.8
429 ± 0.4 , relatively consistent with published values (*e.g.*, Gunn and Mistry, 1970; Simon and Ibrahim,
430 1990; Vandenhove et al., 2009; Mrdakovic Popic et al., 2020). The mean root-to-stem and stem-to-
431 shoot ratios are 2.7 ± 0.3 and 0.8 ± 0.2 , respectively. The stem-to-shoot ratio is slightly smaller for
432 the B subset (0.7 ± 0.1 ; $n = 7$) than for the A subset (1.0 ± 0.2 ; $n = 7$). ^{226}Ra concentration is indeed
433 larger in leaves than in branches or stems for some plants collected in metamorphic context (Saint-
434 Yrieix-la-Perche), and for several plants in carbonaceous and sedimentary context (Pech Merle,
435 Anduze, Vaugrigneuse, and Chambon-la-Forêt). Thus, ^{226}Ra concentration and R_{SP} values decrease
436 in the following order: roots > bark > branches and stems \approx leaves (> flowers > fruits).

437 Samples from the same plant compartment, but collected on different nearby individuals,
438 show mean R_{SP} dispersion of $32 \pm 6 \%$ (Tables 2 and 3). Several nearby top soil samples were
439 collected at some sites. While EC_{Ra} values of soil samples are relatively homogeneous, indicating
440 our soil samples are representative of the local soil for each site, as detailed above, dispersion of
441 soil EC_{Ra} values amounts to $26 \pm 8 \%$ in the Sapine watershed (A subset; $n = 6$), $34 \pm 11 \%$ in
442 Anduze (A subset; $n = 6$), and $27 \pm 7 \%$ in Pech Merle (B subset; $n = 8$). These dispersion values
443 are close to the scarp-scale dispersion of rock EC_{Ra} observed at given sites (from 17 to 69%; Girault
444 and Perrier, 2012b; Girault et al., 2012), suggesting some relation between dispersion of R_{SP} values
445 of nearby individuals and substrate or substratum heterogeneity.

446 In addition, R_{SP} values are relatively similar in the compartments of different plant species
447 growing at the same site. In the Saint-Yrieix leptynite (A subset), a narrow range of R_{SP} values (3–
448 13%) is obtained for the deciduous trees *Quercus robur* and *Castanea sativa*, the shrub *Spartium*
449 *junceum*, and the fern *Pteridium aquilinum* (Table 2 and Fig. 6). In the Kimmeridgian limestone
450 near Puéchabon (B subset), a narrow range of R_{SP} values (0.2–0.8%) is also obtained for the

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451 evergreen tree *Quercus ilex* and the shrubs *Buxus sempervirens* and *Phillyrea latifolia* (Table 3 and
452 Fig. 6). These observations indicate a strong control of the substratum–substrate pair on the R_{SP} of
453 plant beyond the influence of the plant species. Some significant differences also emerge. In the
454 Cévennes granite near Anduze (A subset), mean R_{SP} value obtained for *Quercus robur* (0.13 ± 0.06)
455 is 8 ± 4 times larger than that for *Pinus pinaster* (0.017 ± 0.006). Considering separately the
456 *Quercus* genus (Supplementary Fig. S6), R_{SP} values range from 0.0012 ± 0.0002 to 0.37 ± 0.06 ,
457 with a mean of 0.0134 ± 0.0005 ($n = 40$). The *Quercus* genus yields a large R_{SP} difference of a
458 factor of 23 ± 2 between the A subset (0.122 ± 0.006 ; $n = 12$) and the B subset (0.0052 ± 0.0003 ; n
459 $= 28$), suggesting that the substratum control on R_{SP} is preponderant for a given plant genus. These
460 observations indicate that oak trees tend to concentrate radium more than other trees or plants.

461 Other samples were also considered: market vegetables, mosses, mushrooms and algae. The
462 EC_{Ra} values of market vegetables ($n = 15$), for which we do not have the associated soil sample,
463 range from 0.05 ± 0.02 to 0.82 ± 0.04 Bq kg⁻¹, with a mean of 0.35 ± 0.08 Bq kg⁻¹, and the mean
464 (min–max) calculated bulk ²²⁶Ra concentration yields 0.41 ± 0.09 (0.05–0.95) Bq kg⁻¹, consistent
465 with other reported values (e.g., Lauria et al., 2009). The EC_{Ra} values of mosses, sampled on the
466 ground and on tree bark, are heterogeneous and depend on the substratum, with larger values for the
467 A subset (mean: 39 ± 25 Bq kg⁻¹; $n = 4$) than for the B subset (1.5 ± 0.3 Bq kg⁻¹; $n = 1$). Their
468 respective R_{SP} values are also significantly larger for the A subset (mean: 0.26 ± 0.14) than for the
469 B subset (0.017 ± 0.005). Our R_{SP} values for mosses in the A subset are consistent with reported
470 values in metamorphic context in Serbia (mean: 0.29 ± 0.02 ; $n = 42$; Dragović et al., 2010) and in
471 granitic gneiss context of the Fen Complex, Norway (mean: 0.27 ± 0.14 ; $n = 5–14$; Mrdakovic
472 Popic et al., 2020), and in the B subset with reported values in silico-carbonate context, Norway
473 (mean: 0.036 ± 0.010 ; $n = 5–14$; Mrdakovic Popic et al., 2020). However, compared with vascular
474 plants, mosses have a larger efficiency to take elements directly by precipitation and airborne
475 deposition from the surrounding air (e.g., Uğur et al., 2003; Krmar et al., 2013; Čučulović et al.,

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476 2016; Wattanavatee et al., 2017). Mosses thus may absorb ^{226}Ra contained for example in the soil
477 dust or transported by aerosols, making the contribution of the substratum delicate to assess reliably
478 without additional data.

479 The EC_{Ra} values of mushrooms, all sampled in the Mont-Lozère granite in the Sapine
480 watershed (A subset), are heterogeneous, ranging from 0.5 ± 0.3 to $11.1 \pm 0.6 \text{ Bq kg}^{-1}$, with a mean
481 of $5 \pm 2 \text{ Bq kg}^{-1}$ ($n = 4$). Their mean (min–max) R_{SP} value gives 0.12 ± 0.05 (0.005–0.26), which
482 appears compatible with published R_{SP} data for fungi (mean: 0.08 ± 0.02 ; $n = 5$; Kirchner and
483 Daillant, 1998). Finally, a sample of filamentous algae was collected in flowing water from a lead
484 and zinc mine in Nepal. Using the dissolved ^{226}Ra concentration in water ($20 \pm 15 \text{ mBq L}^{-1}$; Perrier
485 et al., 2016a; Girault et al., 2018) and the EC_{Ra} value of the algae sample ($12.8 \pm 0.9 \text{ Bq kg}^{-1}$), we
486 obtain a large value of ^{226}Ra absorption factor by the algae (740 ± 560), similar to other reported
487 absorption factors of filamentous algae near operating mines in India (Jha et al., 2010).

488

489 *4.3. Differences in emanation coefficients cannot explain the substratum effect on R_{SP}*

490

491 In our study, we have considered a normalization of our EC_{Ra} data by reasonably
492 representative ^{222}Rn emanation coefficient values for soils (E_{S}) and plants (E_{P}) and to calculate the
493 ^{226}Ra soil-to-plant transfer ratio (R_{SP}), a parameter commonly used in the literature. As clearly
494 stated above, these mean E values were chosen based on the results of thorough laboratory
495 experiments using the same method (Perrier et al., 2018). In the following, we discuss the choice of
496 such representative E_{S} and E_{P} values, opening perspectives for future studies.

497 Concerning the ^{222}Rn emanation coefficient for soils, E_{S} , we used a value obtained for sands
498 (0.242 ± 0.035 ; Perrier et al., 2018), as our soils are actually sandy soils, especially at the Sapine
499 site located in Mont-Lozère area (granitic arena; A subset), but also in the sedimentary regions at
500 the Vaugrigneuse site (Fontainebleau sands near Paris; B subset) and at the Chambon-la-Forêt site

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501 (sandy loam near Orléans; B subset). While some systematic differences could be present between
502 soils and sands, the current data do not show any significant difference. Considering the work by
503 Markkanen and Arvela (1992), which is by far the most comprehensive paper on E_S values with
504 more than 400 Finnish soils analyzed, no systematic significant differences in E_S was found
505 between different soil types (clay, silt, sand, gravel and till), with a mean varying from 0.17 to 0.24
506 and 50% of data in the range 0.11 to 0.31, hence compatible with our mean reference E_S value
507 obtained for sands (0.242 ± 0.035). In the Sakoda et al. (2011) review and in all other publications
508 with sufficient numbers of experimental data, the range of variation of E_S is large. This is indeed
509 expected, since the distribution of such data is log-normal and trace element concentrations are
510 affected by large sample-to-sample dispersion (*e.g.*, Bossew, 2010). However, the full min–max
511 range does not give any information on systematic effects on the mean value, which is the only
512 aspect in which we are interested for our current problem. One important point, nevertheless, as
513 detailed below, concerns whether or not we can expect a systematic difference in E_S between our A
514 and B subsets, and whether it may explain the 12 ± 1 times difference in R_{SP} between them.

515 Concerning the ^{222}Rn emanation coefficient for plants, E_P , we used a mean value obtained
516 from different plants and different compartments of a given plant (0.862 ± 0.044 ; Perrier et al.,
517 2018). To our knowledge, the E_P parameter was completely unknown before this study, and thus
518 would be welcome to be refined in detail for various plant samples and plant compartments. Given
519 the small range of variation of E_P based on our experimental data (0.82–0.95; Perrier et al., 2018),
520 an effect of a systematic difference of E_P that may explain the 12 ± 1 times difference in R_{SP}
521 between the A and B subsets can safely be ruled out.

522 The soils overlying the two geological contexts A and B considered in our study have
523 different properties and characteristics. Although the effect of the soil type on the value of E_S is
524 poorly known, some results are nonetheless available. In a study in which five types of soils
525 overlying different lithologies (limestone, gray shale, red shale, gneiss, and sandstone) were

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526 analyzed for their respective E_S values (Luetzelshwab et al., 1989), no significant systematic
527 difference was observed between E_S over gneiss (mean: 0.340 ± 0.039) and limestone (mean: 0.370
528 ± 0.036), with also similar range of values. Similarly, no major systematic difference in E_S related
529 to substratum effects emerges from the Sakoda et al. (2011) review. Consequently, to date, no
530 significant difference in E_S values has been reported between soils overlying rocks of our A and B
531 subsets, hence ruling out any large systematic effect able to explain the obtained large factor of $12 \pm$
532 1. However, this aspect is under-constrained in the literature and would need dedicated experiments.

533 Another aspect we could develop here is the potential effect of soil pH on the ^{222}Rn
534 emanation coefficients of our soils. The values of water pH of our soils (Supp. Mat. Fig. S7),
535 ranging from 3.93 ± 0.01 to 6.39 ± 0.01 in our nine investigated sites, suggest acidic soils and are
536 compatible with the published map of French soils (GIS Sol, 2011). For one of the site (the Sapine
537 in Mont-Lozère; A subset), our mean pH value (4.27 ± 0.27 ; $n = 3$) is consistent with the detailed
538 results already published (Didon-Lescot, 1996). The obtained pH values are on average slightly
539 larger for the B subset (5.82 ± 0.26 ; $n = 11$) than for the A subset (4.71 ± 0.27 ; $n = 7$), as expected.
540 In a recently published paper, Thu et al. (2020) present values of E_S together with the measured
541 water pH of the soil and suggest, despite a large dispersion, a slight variation of E_S versus pH.
542 Using the fit we inferred from Thu et al. (2020) data ($E_S=0.07+0.036\text{pH}$), we get corresponding
543 values of $E_S = 0.24 \pm 0.01$ for the A subset and $E_S = 0.28 \pm 0.01$ for the B subset. Thus, we can
544 expect a difference of E_S between the A and B subsets of $14 \pm 1 \%$, which is small compared with
545 the observed difference in R_{SP} of a factor of 12 ± 1 . Consequently, even when this recent work is
546 taken into account, there is so far no evidence for a sufficient systematic difference of the mean
547 value of E_S between the A and B subsets, and the difference in R_{SP} between them cannot be
548 attributed to a systematic difference in the ^{222}Rn emanation coefficient of soils or plants.

549

550 4.4. Radium uptake as a function of the concentration in the soil

551

552 As in the case of uranium (*e.g.*, Sheppard and Everden, 1988a), a power-law relationship has
553 been proposed between the bulk ^{226}Ra concentration of plant and that of soil, referred to as the
554 ‘linearity hypothesis’, for high ^{226}Ra levels in contaminated areas (Simon and Ibrahim, 1990;
555 Blanco Rodríguez et al., 2002, 2006). Our data set, although obtained in non-contaminated areas
556 with comparatively low ^{226}Ra levels, seems to also follow, to first order, a power-law relationship
557 (dash-dot curve in Fig. 4b). However, several data clearly depart from the linearity hypothesis (Fig.
558 4b), which may suggest the presence of competition between calcium and radium, for example in
559 calcareous context (B subset), as discussed above. More interestingly, the relationship built on a
560 data set obtained for high ^{226}Ra levels, and considered to better reproduce the uptake behavior of
561 calcium and other alkaline earth elements, appears to account for our whole data set (Simon and
562 Ibrahim, 1987, 1990). Indeed, for the relationship between bulk ^{226}Ra concentration in soil and that
563 in plant (solid line in Fig. 4b), and R_{SP} values (solid line in Fig. 4c), our data set is consistent with
564 this empirical fit, indicating its validity for low ^{226}Ra levels. The difference in Fig. 4c between R_{SP}
565 obtained in granitic and metamorphic context (A subset) and R_{SP} obtained in calcareous and
566 sedimentary context (B subset) then reflects the fact that R_{SP} follows a universal function of the
567 ^{226}Ra concentration of the substratum.

568 Our study on ^{226}Ra uptake by plants may be of potential interest to the uptake of other alkali
569 earth elements (Ca, Ba, Sr, and Mg). As an example, Sr, highly toxic to living animals, like Ra, is
570 also in competition with Ca through root uptake, and has been found to affect plant physiology at
571 various levels (Moyen and Roblin, 2010). Thus, our methodological and substratum comparison
572 approach may be a significant contribution not only to soil-to-plant transfer of radium, but also to
573 the radioecology of alkali earth elements.

574

575 **5. Conclusion**

576

577 We have shown that measuring effective ^{226}Ra concentration (EC_{Ra}) can be useful to
578 quantify the ^{226}Ra uptake by the biosphere from soil or water in various environments. We have
579 found that, in non-contaminated areas, R_{SP} values are heterogeneous but reveal an important
580 influence by the substratum, with about an order of magnitude larger R_{SP} values in granitic and
581 metamorphic context than in calcareous and sedimentary context. This difference may likely be due
582 to the competition between calcium and radium, which is being evidenced for the first time in non-
583 contaminated natural environments. In addition, some significant differences emerge between tree
584 species. One first consequence is that results obtained in one geological context cannot be
585 transferred without caution to another context. Another consequence is that the radium flux into the
586 biosphere is larger in granitic context. This may also be the case for strontium, barium, or heavy
587 metals, but this would require confirmation by dedicated studies.

588 Before definite conclusions can be drawn for potential applications such as the
589 characterization of present and past pollution and phytoremediation, the differences observed in the
590 radium uptake need further confirmation and the processes need to be clarified. In particular, the
591 R_{SP} distributions suggesting that soil-to-plant transfer of ^{226}Ra and other radionuclides could be log-
592 normal, so far not considered in most radioecological models (e.g., Whicker et al., 1999; Tuovinen
593 et al., 2016), would need to be studied in details at control sites. A systematic comparison of our
594 effective ^{226}Ra concentration approach (giving effective concentrations) with other well-established
595 methods such as gamma and alpha spectrometry, liquid scintillation counting, and mass
596 spectrometry (giving total concentrations) should be considered in the future. The EC_{Ra} method
597 may also be used to assess the ^{226}Ra fraction bioavailable for terrestrial plants, because it might be
598 close to the ^{226}Ra accessible to the pore space; thus EC_{Ra} could qualify as an appropriate proxy the
599 ^{226}Ra concentration relevant for biosphere uptake and plant ecology, therefore a useful complement

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600 to other approaches. This proposal, which can only be considered as a hypothesis at the moment,
601 could be tested using leaching experiments on ^{226}Ra -rich materials in the laboratory, following
602 already established protocols (*e.g.*, Rihs et al., 2011; Georgiev et al., 2014; Chautard et al., 2020).
603 Such an ambitious program would need to be designed carefully, with an adequate selection of the
604 sites and of the considered plant species. The results obtained in this study provide some initial
605 information that would need to be considerably expanded.

606 Our EC_{Ra} measurement method, cost-effective and easy to implement, may be used at a
607 larger scale to determine R_{SP} value in natural environments characterized by low ^{226}Ra levels and in
608 uranium- and radium-contaminated environments as well (Sheppard et al., 2006; Vandenhove et al.,
609 2009). In addition, to clarify the processes, EC_{Ra} measurements can be incorporated in controlled
610 conditions in the laboratory to study the factors influencing R_{SP} in a systematic manner
611 (Vandenhove et al., 2005; Vandenhove and Van Hees, 2007; Nezami et al., 2016). Given the
612 relatively cost-effective method of radon emanation, which allows large number of data in
613 numerous environments, the uptake of radium by plants in various environments can become an
614 essential tool to study the matter fluxes in the CZ, and reveal, through the pathways of an efficient
615 tracer, the mechanisms to protect or rehabilitate the most fragile parts of the living Earth.

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617

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865 **Tables caption**

866

867 **Table 1.** Statistics of effective ^{226}Ra concentration (EC_{Ra}) of soil and plant samples, and inferred
868 ^{226}Ra soil-to-plant transfer ratios (R_{SP}). All experimental uncertainties are defined at one standard
869 deviation, 68% confidence level (CL).

870

871 **Table 2.** Statistics of effective ^{226}Ra concentration (EC_{Ra}) of soil and plant samples, and inferred
872 ^{226}Ra soil-to-plant transfer ratios (R_{SP}) from different compartments of plant species growing in
873 granitic and metamorphic context (A subset).

874

875 **Table 3.** Statistics of effective radium-226 concentration (EC_{Ra}) of soil and plant samples, and
876 inferred ^{226}Ra soil-to-plant transfer ratios (R_{SP}) from different compartments of plant species
877 growing in calcareous and sedimentary context (B subset).

878 **Figures caption**

879

880 **Fig. 1.** Location of sampling sites in France with simplified geological domains (modified from
881 BRGM, France).

882

883 **Fig. 2.** Relative experimental uncertainty (in %) as a function of EC_{Ra} (in $Bq\ kg^{-1}$) for our plant
884 measurements. Our other measurements are plotted for comparison. The symbol color stands for the
885 sample mass used in the accumulation experiments.

886

887 **Fig. 3.** Distributions of EC_{Ra} values of (a) rocks ($n = 2015$), (b) soils and sediments ($n = 2070$), and
888 (c) plants ($n = 174$). The data from rocks, including samples from Nepal (1128), France (477), and
889 other locations, were partially presented by Girault and Perrier (2019). The data from soils and
890 sediments, including samples from France (1400), Nepal (432), and other locations, and from
891 plants, were partially presented by Perrier et al. (2018). Cumulated distributions (scale on the right
892 hand side) are shown as solid black curves and geometric mean values as vertical dashed black
893 lines. The dashed gray curves represent the log-normal distribution with mean and RMS of $1.9\ Bq$
894 kg^{-1} and 6.0 in (a), and $7.1\ Bq\ kg^{-1}$ and 1.8 in (b), respectively. In (b), the distribution of EC_{Ra}
895 values of top soils nearby plant samples ($n = 24$) is shown in red. In (c), the distribution of EC_{Ra}
896 values of plants for which we have a nearby top soil sample ($n = 108$) is also shown in red.

897

898 **Fig. 4.** (a) Plant EC_{Ra} as a function of EC_{Ra} of the closest top soil sample. (b) Plant EC_{Ra} divided by
899 the emanation coefficient of plants E_P (*i.e.*, bulk plant C_{Ra}) as a function of the closest top soil
900 sample EC_{Ra} divided by the emanation coefficient of soil E_S (*i.e.*, bulk soil C_{Ra}). (c) ^{226}Ra soil-to-
901 plant transfer ratio (R_{SP}) as a function of the bulk soil C_{Ra} . Our whole data set is plotted separately
902 for plants growing in granitic and metamorphic context (A subset; in red) and in calcareous and

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903 sedimentary context (B subset; in blue). The closest soil corresponds to a nearby top soil sample
904 available at a distance of less than 100 m from the given plant sample. The data are represented as
905 diamonds and the means as circles. In (a) and (b), the dashed line represents the radioactive
906 equilibrium between plant and soil samples. In (b), the dash-dot curve corresponds to a power-law
907 regression fit ($EC_{Ra}/E_P = 0.00344(EC_{Ra}/E_S)^{1.37}$; $R^2 = 0.32$). In (b) and (c), the solid black curve
908 represents the exponential relation established by Simon and Ibrahim (1990) for high ^{226}Ra levels in
909 soil: in (b) $EC_{Ra}/E_P = 0.04(1 - \exp(-5.1EC_{Ra}/E_S)) + 0.028EC_{Ra}/E_S$, and in (c) $R_{SP} = 0.04/(EC_{Ra}/E_S)(1 -$
910 $\exp(-5.1EC_{Ra}/E_S)) + 0.028$. In (c), the grey line, $R_{SP} = 1.24 \times 10^5 (EC_{Ra}/E_S)^{-3.43}$, separates the A and B
911 subsets.

912

913 **Fig. 5.** Distributions of ^{226}Ra soil-to-plant transfer ratio, R_{SP} , for (a) all our plant samples for which
914 we have a nearby top soil sample ($n = 108$), (b) plant samples in the A subset (granitic and
915 metamorphic context; $n = 50$), (c) plant samples in the B subset (calcareous and sedimentary
916 context; $n = 58$), and (d) compiled data from the literature ($n = 844$). The data shown in (a) were
917 partially presented in Perrier et al. (2018). Geometric mean values are shown as vertical dashed
918 black lines. In (d), the dashed gray curve represents the log-normal distribution with mean of 0.045
919 and RMS of 5.0.

920

921 **Fig. 6.** ^{226}Ra soil-to-plant transfer ratio, R_{SP} , expressed in percent, for different compartments of
922 given plant species growing in granitic and metamorphic context (A subset; in red, on the right) and
923 in calcareous and sedimentary context (B subset; in blue, on the left).

Figure 1

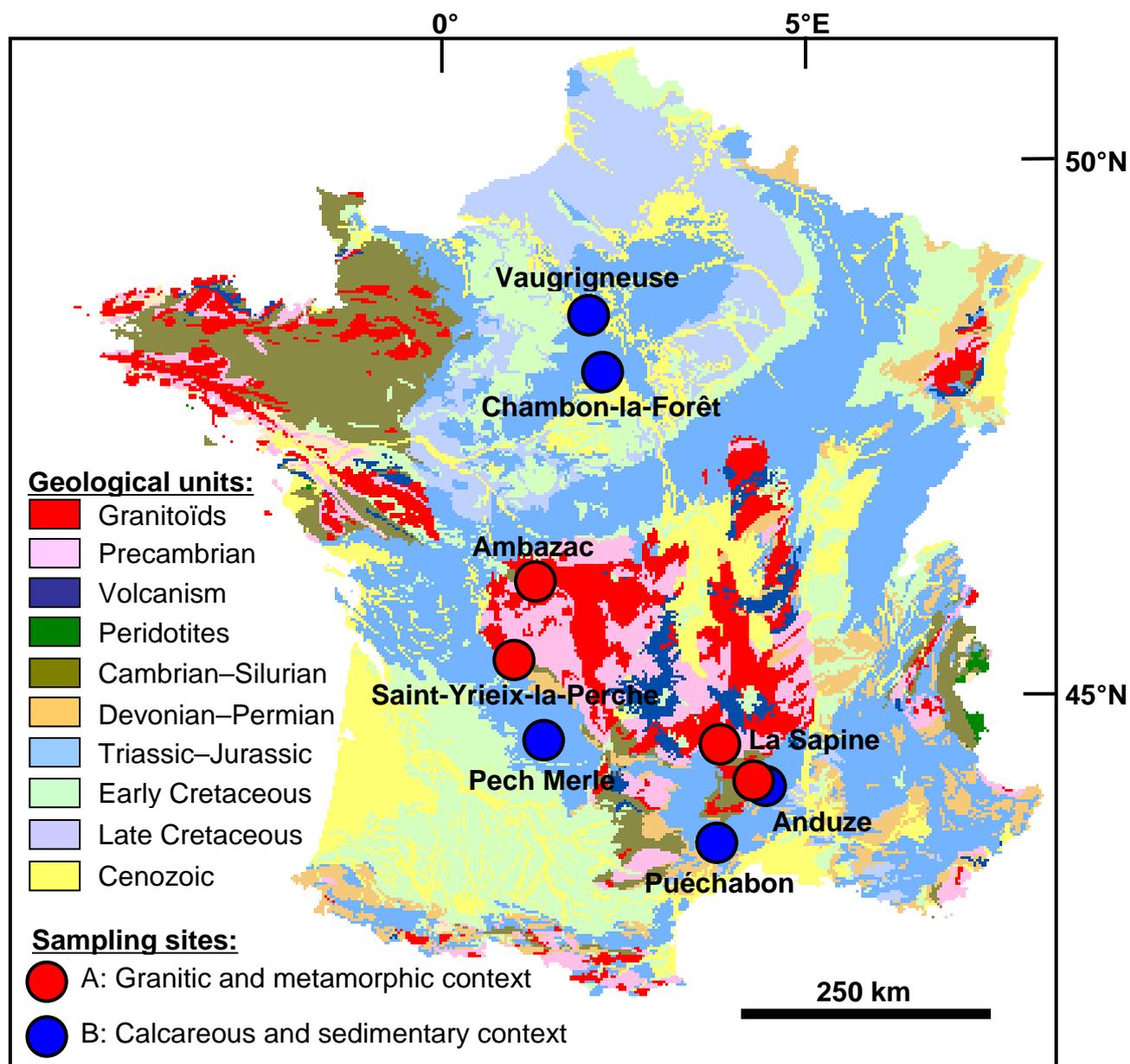


Figure 2

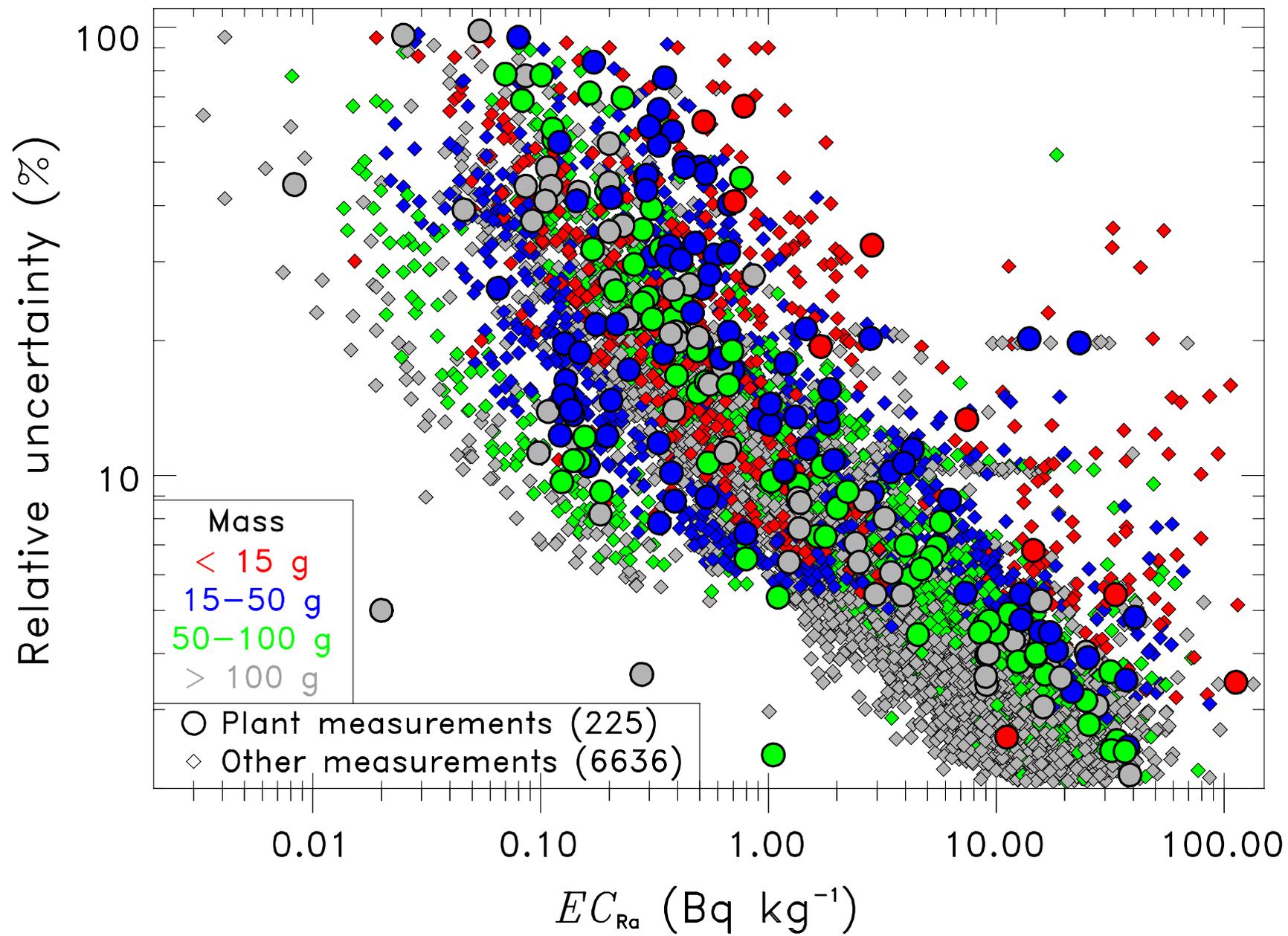


Figure 3

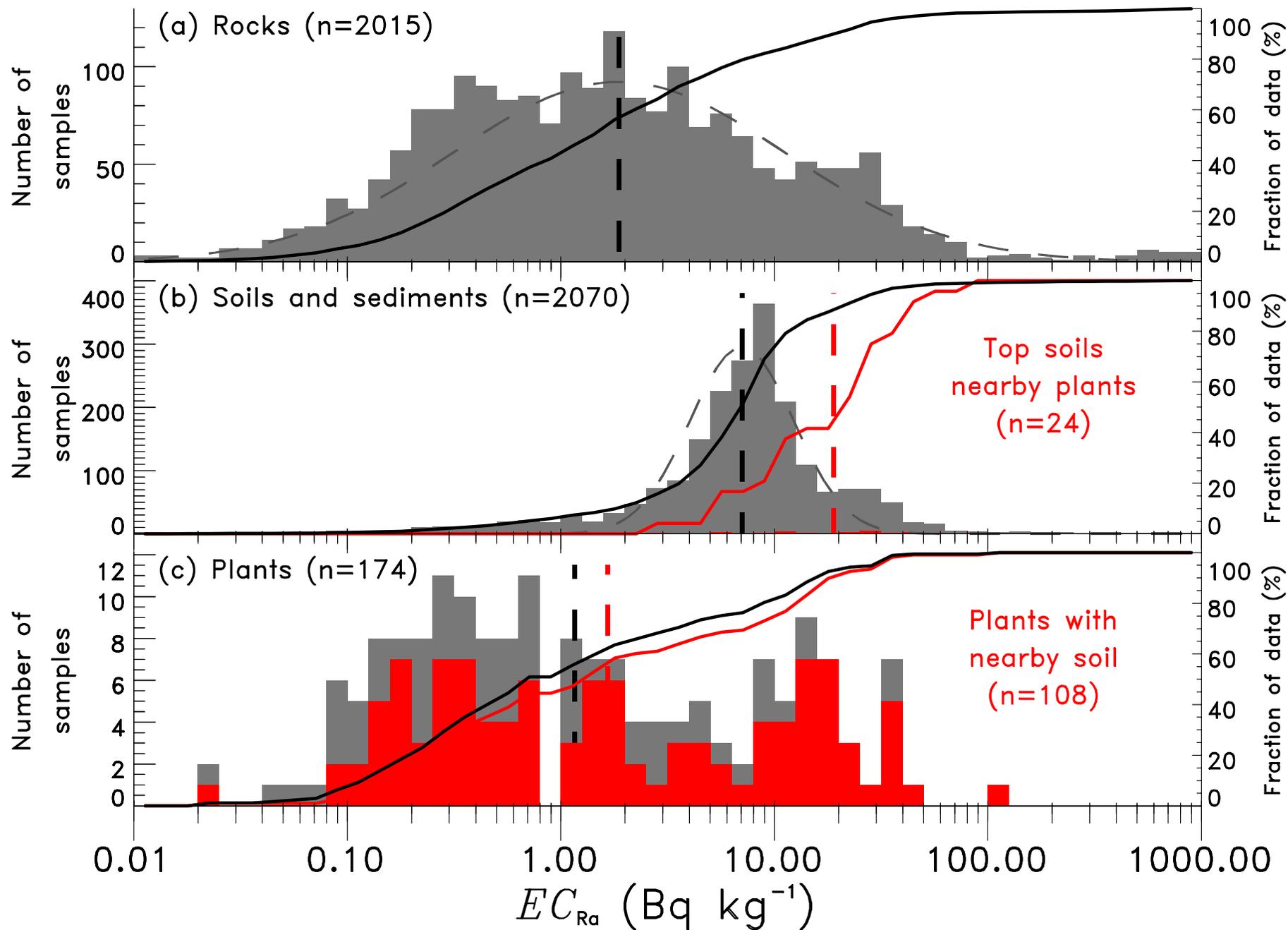


Figure 4

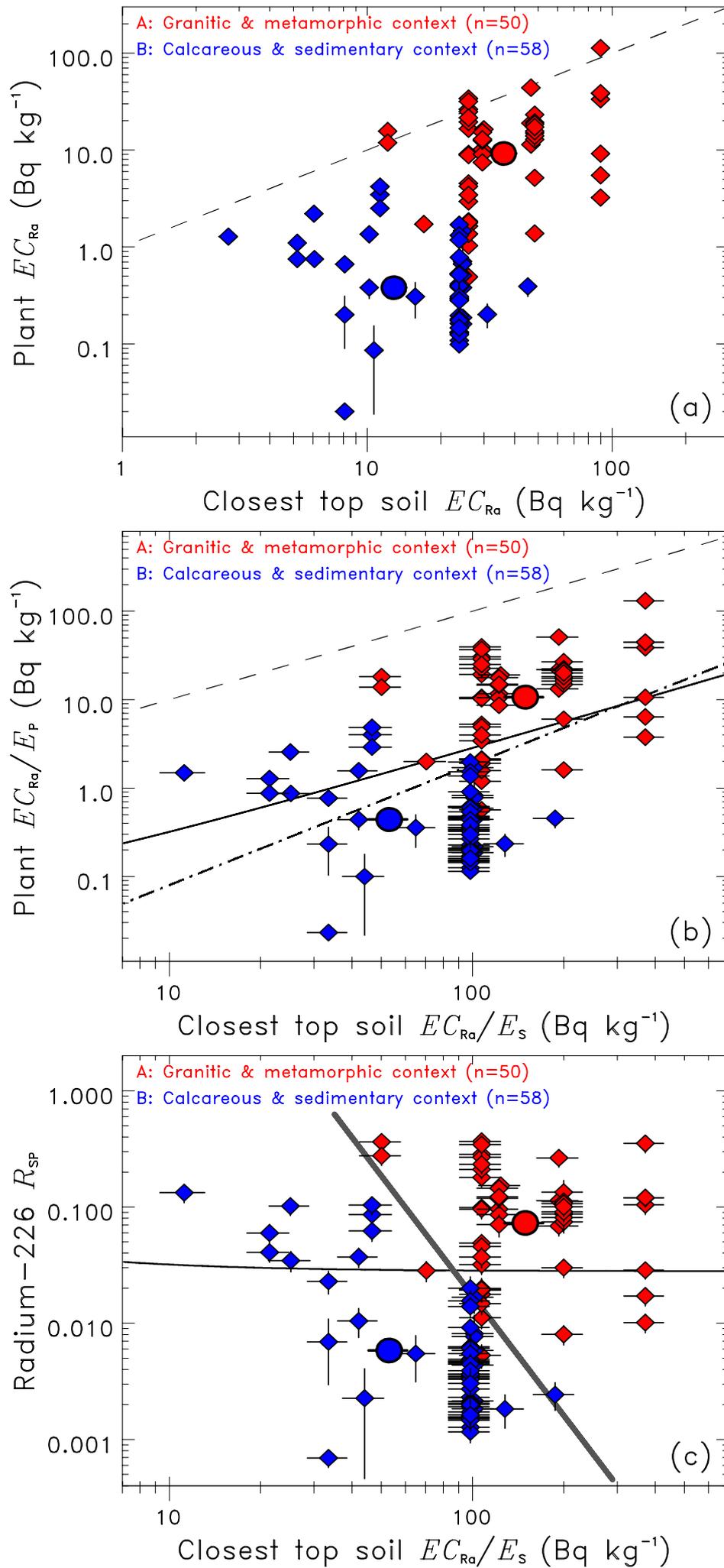


Figure 5

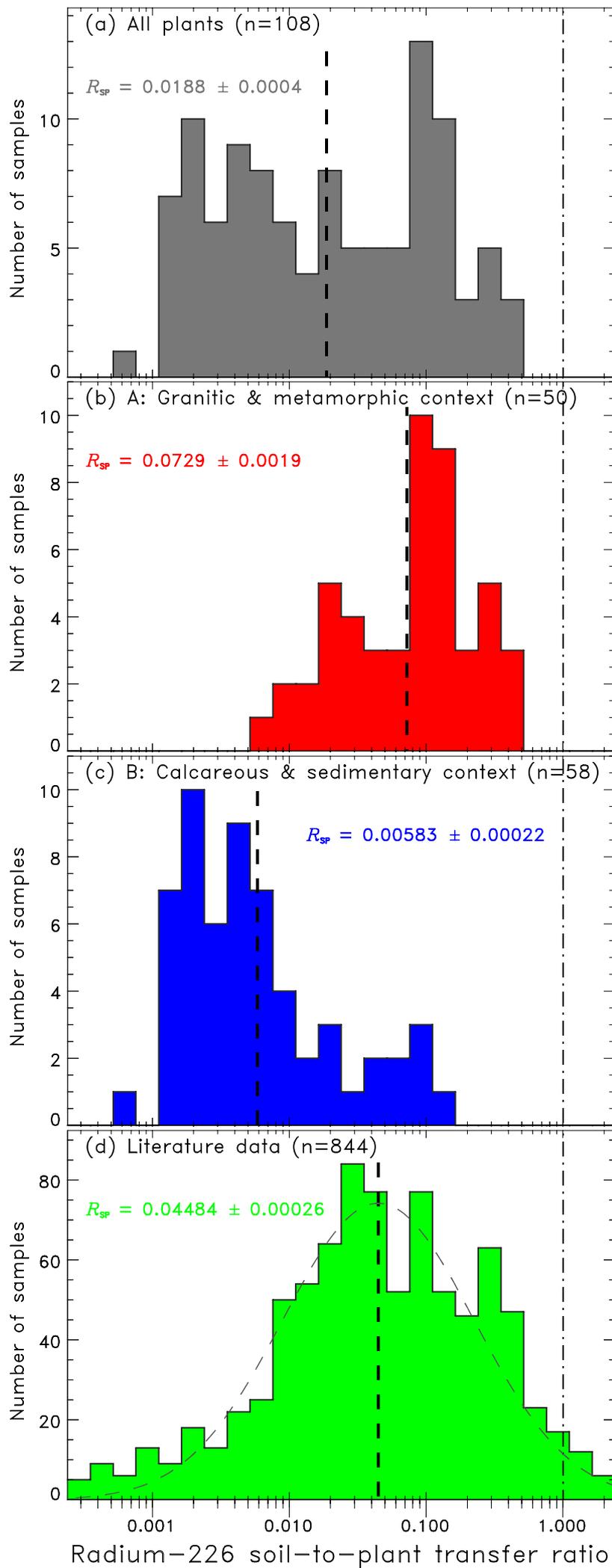
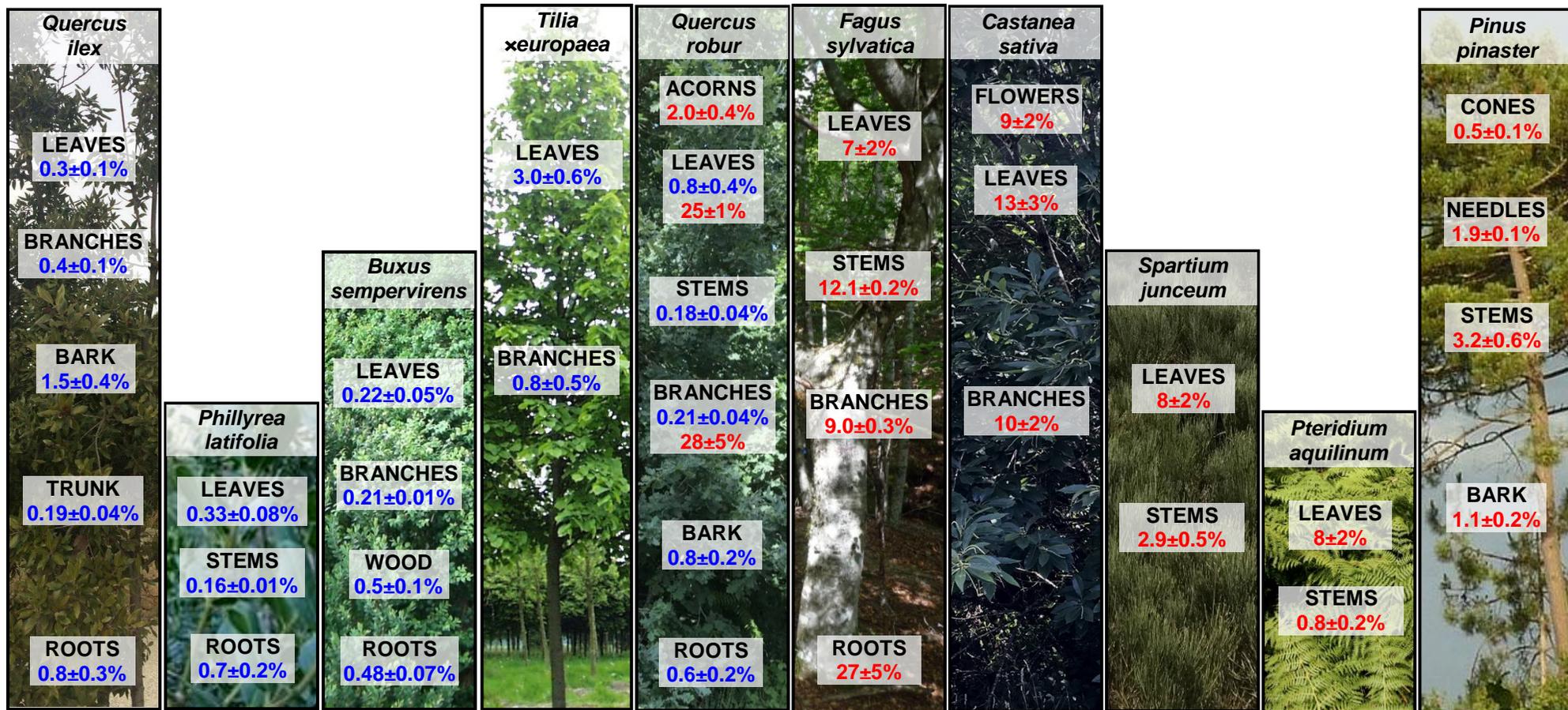


Figure 6



B: CALCAREOUS & SEDIMENTARY CONTEXT

A: GRANITIC & METAMORPHIC CONTEXT

Table 1

Plant samples set	All set	A subset: Granitic & metamorphic context	B subset: Calcareous & sedimentary context
Soil EC_{Ra} (Bq kg⁻¹)			
Number of samples	24	9	15
Min–Max	2.7–90	12–90	2.7–45
Arithmetic mean	25.6 ± 4.0	41.1 ± 7.4	16.8 ± 3.2
Geometric mean	18.86 ± 0.21	36.12 ± 0.64	12.85 ± 0.18
Median (at 90 % CL)	23.80 ± 0.36	35.8 ± 5.4	11.3 ± 2.2
Plant EC_{Ra} (Bq kg⁻¹)			
Number of samples	108	50	58
Min–Max	0.020–113	0.49–113	0.020–4.18
Arithmetic mean	7.6 ± 1.4	15.8 ± 2.5	0.64 ± 0.11
Geometric mean	1.663 ± 0.029	9.21 ± 0.12	0.380 ± 0.012
Median (at 90 % CL)	1.36 ± 0.01	12.83 ± 0.02	0.380 ± 0.001
<i>R_{sp}</i>			
Min–Max	0.00069–0.37	0.0053–0.37	0.00069–0.13
Arithmetic mean	0.0625 ± 0.0085	0.117 ± 0.015	0.0158 ± 0.0038
Geometric mean	0.01877 ± 0.00044	0.0729 ± 0.0019	0.00583 ± 0.00022
Median (at 90 % CL)	0.01928 ± 0.00011	0.0959 ± 0.0015	0.00455 ± 0.00006

Table 2

Sampling site	Plant species and location	Soil EC_{Ra} (Bq kg ⁻¹)	Plant EC_{Ra} (Bq kg ⁻¹)	²²⁶ Ra soil-to-plant transfer ratio R_{SP}	
				Mean $\pm 1\sigma$	Dispersion $\pm 1\sigma$ (%) (number of samples)
La Sapine, Mont Lozère granite, Lozère, France	<i>Fagus sylvatica</i>	29.6 \pm 1.6			
	Dead leaves		15.25 \pm 0.70	0.145 \pm 0.024	
	Leaves		7.4 \pm 1.1	0.071 \pm 0.015	
	Stems		14.1 \pm 1.1	0.1209 \pm 0.0015	2.5 \pm 1.3 (n = 2)
	Branches		10.5 \pm 1.0	0.0898 \pm 0.0026	5.8 \pm 2.9 (n = 2)
Anduze, Cévennes granite, Gard, France	<i>Quercus robur</i>	25.9 \pm 1.4			
	Acorns		1.78 \pm 0.16	0.0193 \pm 0.0036	
	Dead leaves		4.53 \pm 0.30	0.0491 \pm 0.0086	
	Leaves		19.50 \pm 0.89	0.211 \pm 0.036	
	Branches	23.1 \pm 4.1	0.2506 \pm 0.0075	39 \pm 14 (n = 5)	
	<i>Pinus pinaster</i>	25.9 \pm 1.4			
	Pine cones		0.490 \pm 0.079	0.0053 \pm 0.0012	
	Pine needles		1.760 \pm 0.061	0.01906 \pm 0.00066	6.0 \pm 2.5 (n = 3)
	Stems		2.95 \pm 0.22	0.0320 \pm 0.0057	
	Bark from trunk		1.03 \pm 0.11	0.0112 \pm 0.0022	
Ambazac, Saint-Sylvestre granite, Haute-Vienne, France	<i>Fagus sylvatica</i>	46.6 \pm 2.7			
	Branches		18.87 \pm 0.79	0.114 \pm 0.019	
	Roots		44.0 \pm 1.8	0.265 \pm 0.045	
	<i>Castanea sativa</i>	89.8 \pm 3.4			
	Burrs		9.15 \pm 0.40	0.0551 \pm 0.0094	
Leaves		38.3 \pm 1.5	0.231 \pm 0.039		
Saint-Yrieix-la-Perche, Saint-Yrieix arc leptynite, Haute-Vienne, France	<i>Quercus robur</i>	48.3 \pm 2.5			
	Leaves		18.5 \pm 1.2	0.107 \pm 0.019	
	Branches		15.6 \pm 1.1	0.091 \pm 0.016	
	<i>Castanea sativa</i>	51.9 \pm 2.7			
	Flowers		17.3 \pm 1.2	0.093 \pm 0.016	
	Leaves		23.1 \pm 4.7	0.125 \pm 0.033	
	Branches		19.2 \pm 1.2	0.104 \pm 0.018	
	<i>Spartium junceum</i>	50.1 \pm 2.6			
	Leaves		15.00 \pm 0.96	0.084 \pm 0.015	
	Stems		5.18 \pm 0.43	0.0290 \pm 0.0053	
	<i>Pteridium aquilinum</i>	50.1 \pm 2.6			
	Leaves		14.0 \pm 2.9	0.078 \pm 0.021	
	Stems		1.38 \pm 0.14	0.0077 \pm 0.0015	

Table 3

Sampling site	Plant species and location	Soil EC_{Ra} (Bq kg ⁻¹)	Plant EC_{Ra} (Bq kg ⁻¹)	²²⁶ Ra soil-to-plant transfer ratio R_{SP}	
				Mean $\pm 1\sigma$	Dispersion $\pm 1\sigma$ (%) (number of samples)
Puéchabon, Kimmeridgian limestone, Hérault, France	<i>Quercus ilex</i>	22.6 \pm 1.2			
	Leaves		0.269 \pm 0.073	0.00339 \pm 0.00096	49 \pm 24 (n = 3)
	Branches and stems		0.33 \pm 0.11	0.0042 \pm 0.0015	60 \pm 32 (n = 3)
	Trunk		0.158 \pm 0.032	0.00193 \pm 0.00039	40 \pm 16 (n = 4)
	Bark from trunk		1.18 \pm 0.34	0.0145 \pm 0.0042	50 \pm 25 (n = 3)
	Roots	0.63 \pm 0.28	0.0078 \pm 0.0035	77 \pm 47 (n = 3)	
	<i>Phillyrea latifolia</i>	22.6 \pm 1.2			
	Leaves		0.262 \pm 0.057	0.00328 \pm 0.00075	33 \pm 15 (n = 3)
	Stems		0.1309 \pm 0.0039	0.001628 \pm 0.000048	5.1 \pm 2.1 (n = 3)
	Roots	0.56 \pm 0.12	0.0071 \pm 0.0017	41 \pm 19 (n = 3)	
	<i>Buxus sempervirens</i>	22.6 \pm 1.2			
	Leaves		0.179 \pm 0.039	0.00223 \pm 0.00049	38 \pm 18 (n = 3)
	Branches		0.1747 \pm 0.0057	0.002114 \pm 0.000014	5.8 \pm 2.9 (n = 2)
Wood	0.42 \pm 0.10		0.0049 \pm 0.0015		
Roots	0.382 \pm 0.051	0.00478 \pm 0.00071	26 \pm 11 (n = 3)		
Pech Merle, Callovo-Oxfordian limestone, Lot, France	<i>Quercus robur</i>	24.5 \pm 1.4			
	Dead leaves		0.381 \pm 0.038	0.00436 \pm 0.00083	
	Leaves		0.71 \pm 0.29	0.0081 \pm 0.0036	
	Stems		0.160 \pm 0.019	0.00184 \pm 0.00037	
	Branches		0.187 \pm 0.014	0.00214 \pm 0.00039	
	Bark from trunk		0.67 \pm 0.14	0.0077 \pm 0.0021	
Roots	0.51 \pm 0.10	0.0058 \pm 0.0015			
Anduze, Triassic gypsum & Oxfordian limestone, Gard, France	<i>Tilia xeuropaea</i>	7.10 \pm 0.71			
	Dead leaves		0.66 \pm 0.10	0.0262 \pm 0.0063	
	Leaves		0.750 \pm 0.085	0.0297 \pm 0.0064	
Branches	0.20 \pm 0.11	0.0079 \pm 0.0046			
Vaugrigneuse, Stampian Fontainebleau sand, Essonne, France	<i>Quercus robur</i>	5.40 \pm 0.34			
	Leaves		1.10 \pm 0.081	0.057 \pm 0.010	
Branches	0.753 \pm 0.052	0.0392 \pm 0.0070			
Chambon-la-Forêt, Burdigalian sand and marls, Loiret, France	<i>Quercus robur</i>	7.10 \pm 0.48			
	Leaves		3.46 \pm 0.27	0.137 \pm 0.025	
	Stems		4.18 \pm 0.23	0.165 \pm 0.029	
Branches	2.50 \pm 0.20	0.099 \pm 0.018			

Graphical Abstract

