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Substratum influences uptake of radium-226 by plants

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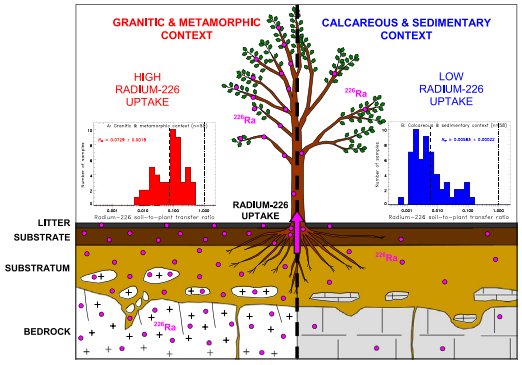
HIGHLIGHTS

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

GRAPHICAL ABSTRACT



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ABSTRACT

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

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1. Introduction

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

Ubiquitous in the environment, the alkaline earth element radium is present in all CZ compartments including water, rock, soil, and vegetation. The radium-226 isotope belongs to the uranium-238 decay chain and is an alpha emitter with a half-life of 1600 ± 7 years (Duchemin et al., 1994). Several studies have focused on the uptake of ^{226}Ra by plants, quantifying a transfer factor (or concentration ratio) of ^{226}Ra from the soil to the plant tissues (Simon and Ibrahim, 1990; Sheppard et al., 2006; Vandenhove et al., 2009; Uchida et al., 2009). Predominantly, because of the need to assess the radium content of agricultural products and the risk to the population, researches on the ^{226}Ra uptake by plants have been carried out in ^{226}Ra-contaminated areas (*e.g.*, Simon and Ibrahim, 1990 and references herein), such as former or operating uranium mining and milling sites (Marple, 1980; Vasconcellos et al., 1987; Bettencourt et al., 1988; Ibrahim and Whicker, 1992; Markose et al., 1993; Madruga et al., 2001; Blanco Rodríguez et al., 2002, 2010; Vera Tomé et al., 2002, 2003; Chen et al., 2005; Ryan et al., 2005; Soudek et al., 2007a, 2007b, 2010; Carvalho et al., 2009; Černe et al., 2011; Medley et al., 2013; Hu et al., 2014; Medley and Bollhöfer, 2016; Yan and Luo, 2016), phosphate fertilizer processing complexes (Paul and Pillai, 1986; Martínez-Aguirre and Periáñez, 1998), radium salt factories (Bettencourt et al., 1988), depleted uranium ammunition sites (Popovic et al., 2008), and other industrial units (Paul and Pillai, 1986). These studies have been complemented by experiments on artificially ^{226}Ra-enhanced soils at the laboratory scale in pots or at larger scale in lysimeter and field experiments (Gerzabek et al., 1998; Bunzl and Trautmannsheimer, 1999; Vandenhove et al., 2005; Vandenhove and Van Hees, 2007; Nezami et al., 2016), providing valuable insights for phytoremediation of contaminated areas (Thiry and Van Hees, 2008; Vera Tomé et al., 2008, 2009; Abreu et al., 2014). By contrast, fewer studies have focused on ^{226}Ra uptake by plants in non-contaminated areas or control sites (Sam and Eriksson, 1995; Ham et al., 2001; Karunakara et al., 2003; Pulhani et al., 2005; Popovic et al., 2008; Uchida and Tagami, 2007; da Conceição et al., 2009; Lauria et al., 2009; Dragović et al., 2010; James et al., 2011; Medley et al., 2013; Asaduzzaman et al., 2014; Al-Hamarneh et al., 2016; Mrdakovic Popic et al., 2020). Despite these efforts, the uptake of ^{226}Ra by plants thus remains insufficiently well understood.

The first mechanism of uptake of elements by plants is passive and results from element concentration in groundwater and evapotranspiration. In this mechanism, ^{226}Ra accumulates in leaves and can also be excreted (*e.g.*, Weis and Weis, 2004). In addition, uptake of elements by a living plant is part of the metabolic cycle. In vascular plants, ^{226}Ra uptake takes place through the various steps of the biological processes (*e.g.*, Simon and Ibrahim, 1990): mobility of Ra^{2+} ions including release and diffusion from the solid phase to the soil solution, exchange of available Ra^{2+} ions by sorption/desorption onto the surfaces of roots, transport of Ra^{2+} ions across membranes in the roots, and diffusion and translocation of ^{226}Ra into plant tissues. Among other factors, it has been recognized that radium uptake depends on the presence of other alkaline earth elements of smaller ionic radius such as barium, strontium, calcium, and magnesium. This suggests that the radium uptake by plants decreases as the concentration of other alkaline earth elements in soil increases, and that incorporation by roots can saturate (Nathwani and Phillips, 1979; Marple, 1980; Simon and Ibrahim, 1987). Generally, a bottom-to-top decreasing gradient (*i.e.*, acropetal) of radium concentration has been observed in plant tissues, from roots to stems and from stems to shoots (*e.g.*, Simon and Ibrahim, 1990). As a testing hypothesis, we could consider that the variability in ^{226}Ra uptake by plants may be due to different types of soil (substrate) and substratum, in particular in non-contaminated areas. However, surprisingly, only small differences or no change at all have been evidenced (*e.g.*, Simon and Ibrahim, 1987, 1990; Vera Tomé et al., 2003; Pulhani et al., 2005).

Several methods, such as gamma-ray spectrometry, alpha-particle spectrometry, liquid scintillation counting, and mass spectrometry, are commonly used to measure high ^{226}Ra levels of numerous materials. However, for low ^{226}Ra levels in soil and for plant samples of relatively small mass, such methods generally give large analytical uncertainty and data have remained limited. Thus, to study ^{226}Ra uptake by plants in non-contaminated areas, an alternative technique is desired, able to reach low ^{226}Ra levels for a large amount of samples, in a cost-effective manner. A candidate high-sensitivity technique with well-constrained leakage effects and relatively small uncertainty for low ^{226}Ra levels is available; it is based on radon-222 emanation, as already suggested thirty years ago (Simon and Ibrahim, 1990). Radon-222 is a radioactive gas (half-life 3.8 days) produced by the alpha decay of ^{226}Ra. The probability that a ^{226}Ra atom decays into a ^{222}Rn atom able to escape from a medium is the emanation coefficient E (Tanner, 1964; Nazaroff, 1992). We define the ^{222}Rn emanating power of a given material by the effective ^{226}Ra concentration (EC_{Ra}), *i.e.* the product of E by the bulk ^{226}Ra concentration (C_{Ra}), expressed in Bq kg^{-1} (Stoulos et al., 2004). Based on the accumulation method, EC_{Ra} has been measured in various materials including soils (*e.g.*, Markkanen and Arvela, 1992; Girault et al., 2011; Perrier et al., 2016b), rocks and building materials (*e.g.*, Przylibski, 2000; Righi and Bruzzi, 2006; Hassan et al., 2011; Girault et al., 2012), and more recently plants (Perrier et al., 2018). Lately, this method has been updated with a significantly higher sensitivity, allowing EC_{Ra} measurement of material with small mass (<5 g) and low ^{226}Ra levels (<10^{-14} g g^{-1}) (Girault et al., 2017a; Girault and Perrier, 2019). Measuring EC_{Ra} of plants using this high-sensitivity method is particularly suited in areas characterized by low ^{226}Ra levels.

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

2. Material and method

2.1. Plant and soil samples

A total of 108 plant samples and their associated nearby local soil samples were collected at different sites in France (Fig. 1). The plants

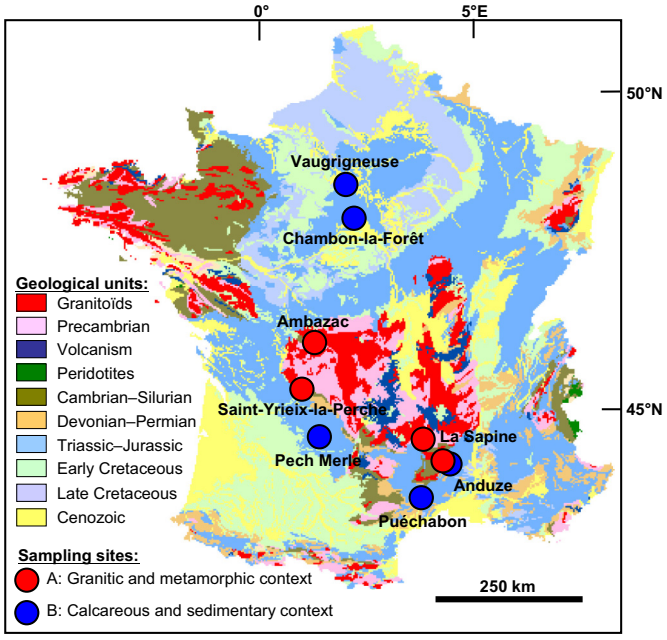

Fig. 1. Location of sampling sites in France with simplified geological domains (modified from BRGM, France).

mainly include deciduous trees (*Quercus robur*, *Fagus sylvatica*, *Castanea sativa*, *Tilia ×europaea*, *Prunus cerasus*, *Ficus carica*, *Fraxinus excelsior*, *Corylus avellana*, *Aesculus hippocastanum*), evergreen trees (*Quercus ilex*, *Pinus pinaster*, *Abies alba*), shrubs (*Buxus sempervirens*, *Phillyrea latifolia*, *Spartium junceum*, *Cytisus oromediterraneus*), and ferns (*Pteridium aquilinum*). Other samples, such as market vegetables ($n = 15$), mosses ($n = 5$), mushrooms ($n = 4$), and algae (n = 1), were also collected.

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

At the selected sites, the radium concentration in groundwater can be considered negligible and the main source of radium is the soil or the regolith. This hypothesis was confirmed at the sites where ^{226}Ra concentration in groundwater could be measured (Perrier et al., 2016a): 4.8 ± 0.4 mBq 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 Pech Merle, 2.7 ± 0.6 mBq L^{-1} ($n = 3$) in Vaugrigneuse, and 15.4 ± 1.6 mBq L^{-1} ($n = 2$) in Chambon-la-Forêt. These values belong to the lower range of ^{226}Ra concentration in groundwater (Girault et al., 2018).

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

2.2. Measurement of water pH of soil samples

Soil pH was determined in the laboratory using the common technique (*e.g.*, Thu et al., 2020). About 20 g of soil and 100 mL of distilled water were placed in a beaker and stirred during 20 to 30 min with a magnetic stirrer. The pH of the suspension solution obtained was measured at about 25 °C using a regularly calibrated pH 211 Microprocessor pH-meter (Hanna Instruments, USA) an hour minimum after stirring to ensure equilibrium. The pH value of a total of 18 representative soils of the two subsets was measured. Absolute experimental uncertainties associated with calibration and reproducibility ranged from 0.01 to 0.02.

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

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

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

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

When ^{226}Ra level was particularly low, a high-sensitivity method was used (Girault et al., 2017a; Girault and Perrier, 2019). In this method, the same protocol was followed, except that air sampling was conducted after a long accumulation time of more than 21 days in order to reach radioactive equilibrium between ^{222}Rn and ^{226}Ra. The number of counts in the flask was then recorded in 10 min interval in

long counting session of 24 h. The background count was determined precisely before sampling during a counting session of 3 days minimum. The final signal was calculated by subtracting from the arithmetic average of the flask count distribution the arithmetic average of the background distribution. Similarly, the effective ^{226}Ra concentration (EC_{Ra}) of the material in the container was determined using Eq. (1). Generally, one or two additional high-sensitivity measurements were performed for samples having an experimental uncertainty larger than about 25% using the first method. For each sample for which significant EC_{Ra} values were obtained using the two methods, or for which EC_{Ra} values were obtained using different subsamples of the same sample, the final retained EC_{Ra} value is a weighted mean accounting for all the available measurements.

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

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

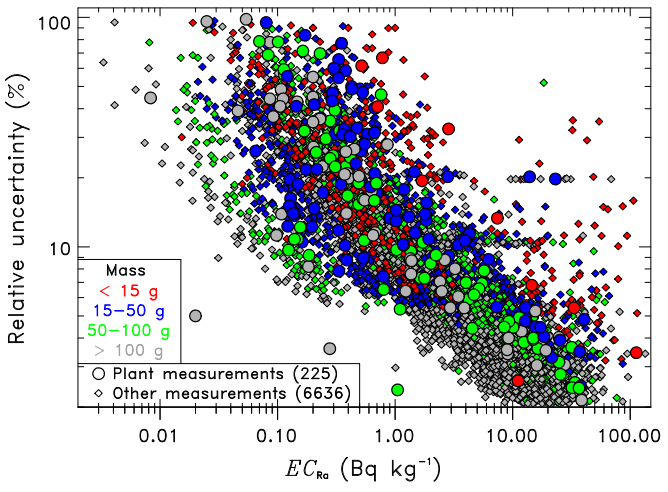

Fig. 2. Relative experimental uncertainty (in %) as a function of EC_{Ra} (in Bq kg^{-1}) for our plant measurements. Our other measurements are plotted for comparison. The symbol color stands for the sample mass used in the accumulation experiments.

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

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

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

The obtained ratio (EC_{RaP}/EC_{RaS}) might be an interesting transfer parameter in itself. Indeed, it is meaningful to normalize the effective ^{226}Ra concentration of the plant sample, which is to a first approximation the ^{226}Ra concentration of the plant sample, as we will show below, to the effective ^{226}Ra concentration of the soil, which represents the amount of ^{226}Ra connected to the pore space, thus available to the root system. Nevertheless, in a second approach and in order to obtain values of the soil-to-plant transfer ratio (R_{SP}), similar to the transfer factor or the concentration ratio commonly used in the literature, we eliminate the mean emanation coefficients from the observed ^{226}Ra plant-to-soil effective concentration ratios. Indeed, because EC_{Ra} gives an effective ^{226}Ra concentration of a given material, dividing EC_{Ra} of a material by its emanation coefficient, E, gives the bulk ^{226}Ra concentration of the material. Here, we used mean representative E values for soils and plants, as inferred from ^{226}Ra doping experiments carried out in the laboratory (Perrier et al., 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 calculated using (Perrier et al., 2018):

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

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

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

3. Results

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

Effective ^{226}Ra concentration (EC_{Ra}) values of plant samples ($n = 108$) range over about five orders of magnitude (Fig. 3c), from 0.020 ± 0.001 to 113 ± 7 Bq kg^{-1}, and with a mean of 1.66 ± 0.03 Bq kg^{-1} (Table 1). The two largest values, 113 ± 7 and 44 ± 2 Bq kg^{-1}, are measured for a moss and roots of *Fagus sylvatica*, respectively, both from the granitic area near Ambazac. The two smallest values, 0.020 ± 0.001 and 0.09 ± 0.07 Bq kg^{-1}, are measured for chestnuts of *Aesculus hippocastanum* and branches of *Prunus cerasus*, respectively, both from the sedimentary area near Anduze. The EC_{Ra} values of the nearby soil samples ($n = 24$) are less scattered (Fig. 3b) with a mean of 18.9 ± 0.2 Bq kg^{-1} (Table 1). While the EC_{Ra} values of plants follow a log-normal distribution with two modes around 0.5 and 15 Bq kg^{-1}, similar to our whole data set of plant EC_{Ra} ($n = 174$) (Fig. 3c), the EC_{Ra} values of soils follow a log-normal distribution with a single mode around the mean (Fig. 3b). Although, to first order, we may expect some relation between EC_{Ra} of plant and EC_{Ra} of soil, the mean value and the general shape of the log-normal distribution of plant EC_{Ra} appear relatively similar to those of rock EC_{Ra} (Fig. 3a), suggesting a possible relation between a given plant and its substratum. Alternatively, the large range of plant EC_{Ra} may also suggest an efficient dispersion process during the transfer of ^{226}Ra from soil to the biosphere.

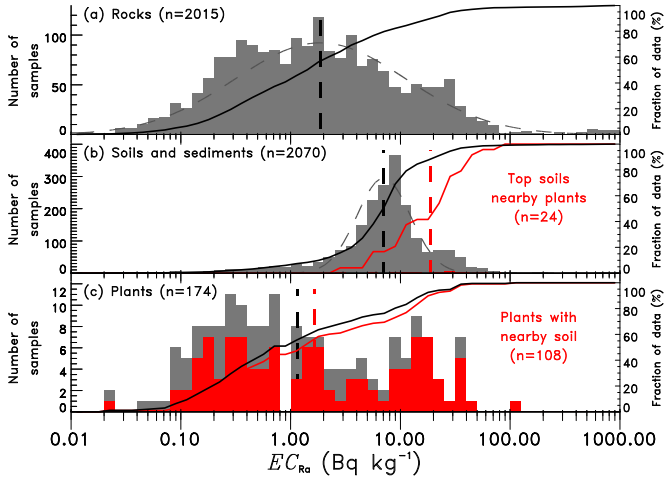

Fig. 3. Distributions of EC_{Ra} values of (a) rocks ($n = 2015$), (b) soils and sediments ($n = 2070$), and (c) plants ($n = 174$). The data from rocks, including samples from Nepal (1128), France (477), and other locations, were partially presented by Girault and Perrier (2019). The data from soils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

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

3.2. Results on the corrected ^{226}Ra soil-to-plant transfer ratio

Normalizing plant EC_{Ra} by emanation coefficient of plants (E_P) and similarly soil EC_{Ra} by 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. 4b). The min–max values of EC_{RaP}/E_P and EC_{RaS}/E_S respectively, corresponding to the bulk ^{226}Ra 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 11–190 Bq kg^{-1} for the B subset, which are consistent with reported ^{226}Ra concentration in plants and soils in similar contexts.

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

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

4. Discussion

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

Our soil-to-plant transfer ratio (R_{SP}) data, although they are inferred from the measurement of effective ^{226}Ra concentration (EC_{Ra}) and emanation coefficient (E) of both soil and plant samples, give compatible values with the transfer factors deduced from the direct measurement of bulk ^{226}Ra concentrations as reported in the literature. Indeed, our

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

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
R_{SP}			
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

mean R_{SP} value, 0.0188 ± 0.0004, thus about 2%, is consistent with mean ^{226}Ra transfer factors reported in the literature for all plants, ranging from about 2% (Vandenhove et al., 2009) to about 5% (Sheppard et al., 2006). Using a compiled data set from published ^{226}Ra transfer factors from the literature (38 articles; $n = 870$), 90% of the R_{SP} values are comprised between 0.0015 and 1. R_{SP} values above 1 are generally obtained with vegetables growing in radium-contaminated environments or in laboratory experiments using radium-rich nutrient solutions (Simon and Ibrahim, 1990 and references herein; Sam and Eriksson, 1995; Martínez-Aguirre and Periáñez, 1998; Vandenhove and Van Hees, 2007; Asaduzzaman et al., 2014; Hu et al., 2014). R_{SP} values below 0.0015 are generally obtained with vegetables, fruits, and cereals growing in radium-contaminated environments or, in a lesser extent, in non-contaminated areas (Simon and Ibrahim, 1990 and references herein; Markose et al., 1993; Ham et al., 2001; Ryan et al., 2005; Uchida and Tagami, 2007; Uchida et al., 2009 and references herein; Černe et al., 2011). Considering a reasonable range of R_{SP} values over five orders of magnitude (from 0.00025 to 2; $n = 844$), these R_{SP} values follow a log-normal distribution with one mode, centered on the mean of 0.0448 ± 0.0003 (Fig. 5d). In the literature, only few R_{SP} values smaller than 0.006 (about 11%) are reported. This small range corresponds to the low R_{SP} values we obtained in calcareous and sedimentary context (B subset). To our knowledge, no study has focused on radium transfer from soil to plants in calcareous and sedimentary context, probably because of the low ^{226}Ra levels that are beyond the detection limit of the commonly used methods (gamma and alpha spectrometry, liquid scintillation counting, and mass spectrometry). This highlights the usefulness of our high-sensitivity EC_{Ra} measurement method, decreasing the detection limit of ^{226}Ra levels in soil and plant samples with reasonable experimental uncertainty.

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

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

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

For some plant species, we collected several plant compartments. When considering separately the two subsets of samples in different substratum contexts, the R_{SP} values obtained for the different compartments of a given plant appear relatively compatible (Fig. 6 and Tables 2 and 3). The acropetal gradient, *i.e.*, a decrease of ^{226}Ra concentration from roots to apex, is generally 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 ± 0.4, relatively consistent with published values (*e.g.*, Gunn and Mistry, 1970; Simon and Ibrahim, 1990; Vandenhove et al., 2009; Mrdakovic Popic et al., 2020). The mean root-to-stem and stem-to-shoot ratios are 2.7 ± 0.3 and 0.8 ± 0.2, respectively. The stem-to-shoot ratio is slightly smaller for 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 larger in leaves than in branches or stems for some plants collected in metamorphic context (Saint-Yrieix-la-Perche), and for several plants in carbonaceous and sedimentary context (Pech Merle, Anduze, Vaugrigneuse, and Chambon-la-Forêt). Thus, ^{226}Ra concentration and R_{SP} values decrease in the following order: roots > bark > branches and stems ≈ leaves (> flowers > fruits).

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

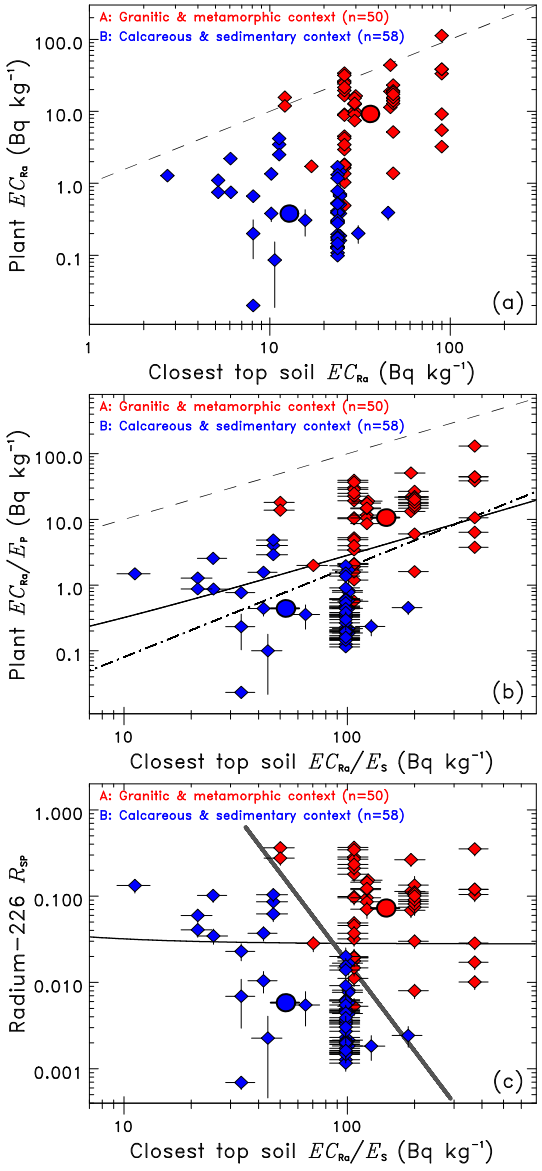

Fig. 4. (a) Plant EC_{Ra} as a function of EC_{Ra} of the closest top soil sample. (b) Plant EC_{Ra} divided by the emanation coefficient of plants E_P (*i.e.*, bulk plant C_{Ra}) as a function of the closest top soil sample EC_{Ra} divided by the emanation coefficient of soil E_S (*i.e.*, bulk soil C_{Ra}). (c) ^{226}Ra soil-to-plant transfer ratio (R_{SP}) as a function of the bulk soil C_{Ra}. Our whole data set is plotted separately for plants growing in granitic and metamorphic context (A subset; in red) and in calcareous and sedimentary context (B subset; in blue). The closest soil corresponds to a nearby top soil sample available at a distance of less than 100 m from the given plant sample. The data are represented as diamonds and the means as circles. In (a) and (b), the dashed line represents the radioactive equilibrium between plant and soil samples. In (b), the dash-dot curve corresponds to a power-law 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 represents the exponential relation established by Simon and Ibrahim (1990) for high ^{226}Ra levels in soil: in (b) $EC_{Ra}/E_P = 0.04(1-\exp(-5.1EC_{Ra}/E_S)) + 0.028EC_{Ra}/E_S$, and in (c) $R_{SP} = 0.04/(EC_{Ra}/E_S)(1-\exp(-5.1EC_{Ra}/E_S)) + 0.028$. In (c), the gray line, $R_{SP} = 1.24 \times 10^5 (EC_{Ra}/E_S)^{-3.43}$, separates the A and B subsets.

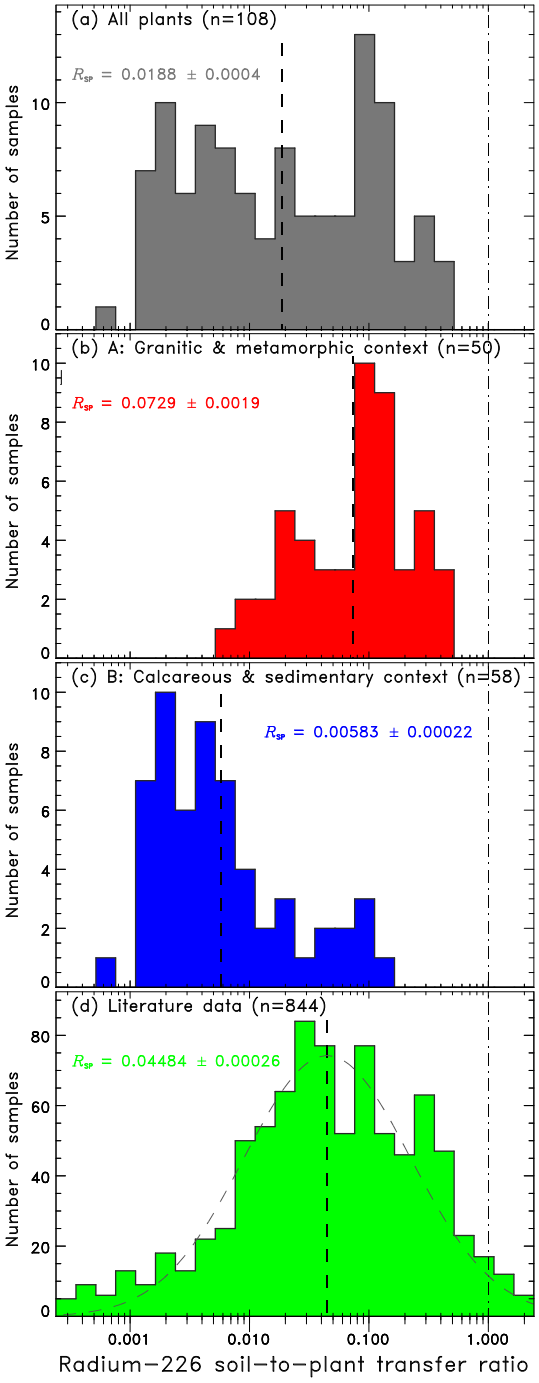

Fig. 5. Distributions of ^{226}Ra soil-to-plant transfer ratio, R_{SP}, for (a) all our plant samples for which we have a nearby top soil sample (n = 108), (b) plant samples in the A subset (granitic and metamorphic context; $n = 50$), (c) plant samples in the B subset (calcareous and sedimentary context;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

In addition, R_{SP} values are relatively similar in the compartments of different plant species growing at the same site. In the Saint-Yrieix leptynite (A subset), a narrow range of R_{SP} values (3–13%) is obtained for the deciduous trees *Quercus robur* and *Castanea sativa*, the shrub *Spartium junceum*, and the fern *Pteridium aquilinum* (Table 2 and Fig. 6). In the Kimmeridgian limestone near Puéchabon (B subset), a narrow range of R_{SP} values (0.2–0.8%) is also obtained for the evergreen tree *Quercus ilex* and the shrubs *Buxus sempervirens* and *Phillyrea latifolia* (Table 3 and Fig. 6). These observations indicate a strong control of the substratum–substrate pair on the R_{SP} of plant beyond the influence of the plant species. Some significant differences also emerge. In the Cévennes granite near Anduze (A subset), mean R_{SP} value obtained for *Quercus robur* (0.13 ± 0.06) is 8 ± 4 times larger than that for *Pinus pinaster* (0.017 ± 0.006). Considering separately the *Quercus* genus

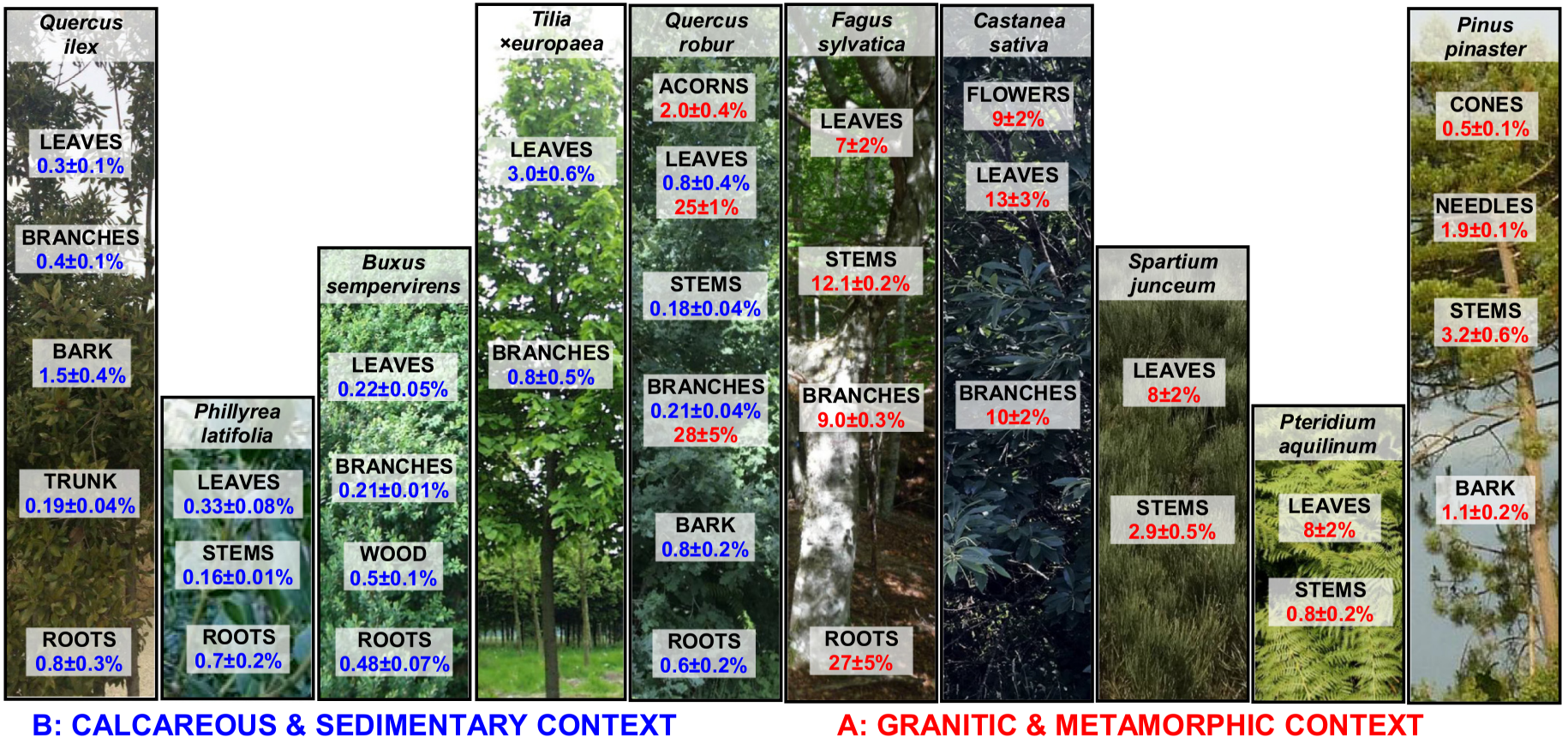

Fig. 6. ^{226}Ra soil-to-plant transfer ratio, R_{SP}, expressed in percent, for different compartments of given plant species growing in granitic and metamorphic context (A subset; in red, on the right) and in calcareous and sedimentary context (B subset; in blue, on the left).

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

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

(Supplementary Fig. S6), R_{SP} values range from 0.0012 ± 0.0002 to 0.37 ± 0.06, with a mean of 0.0134 ± 0.0005 (n = 40). The *Quercus* genus yields a large R_{SP} difference of a factor of 23 ± 2 between the A subset (0.122 ± 0.006; n = 12) and the B subset (0.0052 ± 0.0003; n = 28), suggesting that the substratum control on R_{SP} is preponderant for a given plant genus. These observations indicate that oak trees tend to concentrate radium more than other trees or plants.

Other samples were also considered: market vegetables, mosses, mushrooms and algae. The EC_{Ra} values of market vegetables (n = 15), for which we do not have the associated soil sample, 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 (min–max) calculated bulk ^{226}Ra concentration yields 0.41 ± 0.09 (0.05–0.95) Bq kg^{-1}, consistent with other reported values (*e.g.*, Lauria et al., 2009). The EC_{Ra} values of mosses, sampled on the ground and on tree bark, are heterogeneous and depend on the substratum, with larger values for the 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 respective R_{SP} values are also significantly larger for the A subset (mean: 0.26 ± 0.14) than for the B subset (0.017 ± 0.005). Our R_{SP} values for mosses in the A subset are consistent with reported values in metamorphic context in Serbia (mean: 0.29 ± 0.02; n = 42; Dragović et al., 2010) and in granitic gneiss context of the Fen Complex, Norway (mean: 0.27 ± 0.14; n = 5–14; Mrdakovic Popic et al., 2020), and in the B subset with reported values in silico-carbonate context, Norway (mean: 0.036 ± 0.010; n = 5–14; Mrdakovic Popic et al., 2020). However, compared with vascular plants, mosses have a larger efficiency to take elements directly by precipitation and airborne deposition from the surrounding air (*e.g.*, Uğur et al., 2003; Krmar et al., 2013; Čučulović et al., 2016; Wattanavatee et al., 2017). Mosses thus may absorb ^{226}Ra contained for example in the soil dust or transported by aerosols, making the contribution of the substratum delicate to assess reliably without additional data.

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

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

In our study, we have considered a normalization of our EC_{Ra} data by reasonably representative ^{222}Rn emanation coefficient values for soils (E_S) and plants (E_P) and to calculate the ^{226}Ra soil-to-plant transfer ratio (R_{SP}), a parameter commonly used in the literature. As clearly stated above, these mean E values were chosen based on the results of thorough laboratory experiments using the same method (Perrier

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

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

et al., 2018). In the following, we discuss the choice of such representative E_S and E_P values, opening perspectives for future studies.

Concerning the ^{222}Rn emanation coefficient for soils, E_S, we used a value obtained for sands (0.242 ± 0.035; Perrier et al., 2018), as our soils are actually sandy soils, especially at the Sapine site located in Mont-Lozère area (granitic arena; A subset), but also in the sedimentary regions at the Vaugrigneuse site (Fontainebleau sands near Paris; B subset) and at the Chambon-la-Forêt site (sandy loam near Orléans; B subset). While some systematic differences could be present between soils and sands, the current data do not show any significant difference. Considering the work by Markkanen and Arvela (1992), which is by far the most comprehensive paper on E_S values with more than 400 Finnish soils analyzed, no significant systematic differences in E_S was found between different soil types (clay, silt, sand, gravel and till), with a mean varying from 0.17 to 0.24 and 50% of data in the range 0.11 to 0.31, hence compatible with our mean reference E_S value obtained for sands (0.242 ± 0.035). In the Sakoda et al. (2011) review and in all other publications with sufficient numbers of experimental data, the range of variation of E_S is large. This is indeed expected, since the distribution of such data is log-normal and trace element concentrations are affected by large sample-to-sample dispersion (*e.g.*, Bossew, 2010). However, the full min–max range does not give any information on systematic effects on the mean value, which is the only aspect in which we are interested for our current problem. One important point, nevertheless, as detailed below, concerns whether or not we can expect a systematic difference in E_S between our A and B subsets, and whether it may explain the 12 ± 1 times difference in R_{SP} between them.

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

The soils overlying the two geological contexts A and B considered in our study have different properties and characteristics. Although the effect of the soil type on the value of E_S is poorly known, some results are nonetheless available. In a study in which five types of soils overlying different lithologies (limestone, gray shale, red shale, gneiss, and sandstone) were analyzed for their respective E_S values (Luetzelschwab et al., 1989), no significant systematic difference was observed between E_S over gneiss (mean: 0.340 ± 0.039) and limestone (mean: 0.370 ± 0.036), with also a similar range of values. Similarly, no major systematic difference in E_S related to substratum effects emerges from the Sakoda et al. (2011) review. Consequently, to date, no significant difference in E_S values has been reported between soils overlying rocks of our A and B subsets, hence ruling out any large systematic effect able to explain the obtained large factor of 12 ± 1. However, this aspect is under-constrained in the literature and would need dedicated experiments.

Another aspect we could develop here is the potential effect of soil pH on the ^{222}Rn emanation coefficients of our soils. The values of water pH of our soils (Supp. Mat. Fig. S7), ranging from 3.93 ± 0.01 to

6.39 ± 0.01 in our nine investigated sites, suggest acidic soils and are compatible with the published map of French soils (GIS Sol, 2011). For one of the site (the Sapine in Mont-Lozère; A subset), our mean pH value (4.27 ± 0.27; $n = 3$) is consistent with the detailed results already published (Didon-Lescot, 1996). The obtained pH values are on average slightly larger for the B subset (5.82 ± 0.26; $n = 11$) than for the A subset (4.71 ± 0.27; $n = 7$), as expected. In a recently published paper, Thu et al. (2020) present values of E_S together with the measured water pH of the soil and suggest, despite a large dispersion, a slight variation of E_S versus pH. Using the fit we inferred from Thu et al. (2020) data ($E_S = 0.07 + 0.036$pH), we get corresponding 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 expect a difference of E_S between the A and B subsets of 14 ± 1%, which is small compared with the observed difference in R_{SP} of a factor of 12 ± 1. Consequently, even when this recent work is taken into account, there is so far no evidence for a sufficient systematic difference of the mean value of E_S between the A and B subsets, and the difference in R_{SP} between them cannot be attributed to a systematic difference in the ^{222}Rn emanation coefficient of soils or plants.

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

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

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

5. Conclusion

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

Before definite conclusions can be drawn for potential applications such as the characterization of present and past pollution and phytoremediation, the differences observed in the radium uptake need further confirmation and the processes need to be clarified. In particular, the R_{SP} distributions suggesting that soil-to-plant transfer of ^{226}Ra and other radionuclides could be log-normal, so far not considered in most radioecological models (*e.g.*, Whicker et al., 1999; Tuovinen et al., 2016), would need to be studied in details at control sites. A systematic comparison of our effective ^{226}Ra concentration approach (giving effective concentrations) with other well-established methods such as gamma and alpha spectrometry, liquid scintillation counting, and mass spectrometry (giving total concentrations) should be considered in the future. The EC_{Ra} method may also be used to assess the ^{226}Ra fraction bioavailable for terrestrial plants, because it might be close to the ^{226}Ra accessible to the pore space; thus EC_{Ra} could qualify as an appropriate proxy the ^{226}Ra concentration relevant for biosphere uptake and plant ecology, therefore a useful complement to other approaches. This proposal, which can only be considered as a hypothesis at the moment, could be tested using leaching experiments on ^{226}Ra-rich materials in the laboratory, following already established protocols (*e.g.*, Rihs et al., 2011; Georgiev et al., 2014; Chautard et al., 2020). Such an ambitious program would need to be designed carefully, with an adequate selection of the sites and of the considered plant species. The results obtained in this study provide some initial information that would need to be considerably expanded.

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

CRediT authorship contribution statement

Frédéric Girault: Conceptualization, Methodology, Formal analysis, Investigation, Resources, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Frédéric Perrier:** Conceptualization, Methodology, Investigation, Resources, Writing - original draft, Writing - review & editing. **Jean-Marc Ourcival:** Investigation, Resources. **Roxane Ferry:** Investigation. **Yves Gaudemer:** Visualization. **François Bourges:** Investigation, Resources. **Jean-François Didon-Lescot:** Investigation, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary figures

Supplementary Figs. S1-7 to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.142655.

References

Abrahams, P.W., 2002. Soils: their implications to human health. Sci. Total Environ. 291, 1–32.
Abreu, M.M., Lopes, J., Santos, E.S., Magalhães, M.C.F., 2014. Ecotoxicity evaluation of an amended soil contaminated with uranium and radium using sensitive plants. J. Geochem. Explor. 142, 112–121.
Adler, P.M., Perrier, F., 2009. Radon emanation in partially saturated porous media. Transport Porous Med. 78, 149–159.
Al-Hamarneh, I.F., Alkhomashi, N., Almasoud, F.I., 2016. Study on the radioactivity and soil-to-plant transfer factor of ^{226}Ra, ^{234}U and ^{238}U radionuclides in irrigated farms from the northwestern Saudi Arabia. J. Environ. Radioact. 160, 1–7.
Asaduzzaman, K., Khandaker, M.U., Amin, Y.M., Bradley, D.A., Mahat, R.H., Nor, R.M., 2014. Soil-to-root vegetable transfer factors for ^{226}Ra, ^{232}Th, ^{40}K, and ^{88}Y in Malaysia. J. Environ. Radioact. 135, 120–127.
Banwart, S., 2011. Save our soils. Nature 474, 151–152.
Bettencourt, A.O., Teixeira, M.M.G.R., Elias, M.D.T., Faisca, M.C., 1988. Soil to plant transfer of radium-226. J. Environ. Radioact. 6, 49–60.
Blanco Rodríguez, P., Vera Tomé, F., Lozano, J.C., 2002. About the assumption of linearity in soil-to-plant transfer factors for uranium and thorium isotopes and ^{226}Ra. Sci. Total Environ. 284, 167–175.
Blanco Rodríguez, P., Vera Tomé, F., Pérez Fernández, M., Lozano, J.C., 2006. Linearity assumption in soil-to-plant transfer factors of natural uranium and radium in *Helianthus annuus* L. Sci. Total Environ. 361, 1–7.
Blanco Rodríguez, P., Vera Tomé, F., Lozano, J.C., Pérez Fernández, M.A., 2010. Transfer of ^{238}U, ^{230}Th, ^{226}Ra, and ^{210}Pb from soils to tree and shrub species in a Mediterranean area. Appl. Radiat. Isotopes 68, 1154–1159.
Bossew, P., 2010. Radon: exploring the log-normal mystery. J. Environ. Radioact. 101, 826–834.
Brantley, S.L., Goldhaber, M.B., Ragnarsdottir, K.V., 2007. Crossing disciplines and scales to understand the critical zone. Elements 3, 307–314.
Bunzl, K., Trautmannsheimer, M., 1999. Transfer of ^{238}U, ^{226}Ra and ^{210}Pb from slag-contaminated soils to vegetables under field conditions. Sci. Total Environ. 231, 91–99.
Carini, F., 1999. Radionuclides in plants bearing fruit: an overview. J. Environ. Radioact. 46, 77–97.
Carvalho, F.P., Oliveira, J.M., Neves, M.O., Abreu, M.M., Vicente, E.M., 2009. Soil to plant (*Solanum tuberosum* L.) radionuclide transfer in the vicinity of an old uranium mine. Geochem. Explor. Environ. Anal. 9, 275–278.
Černe, M., Smodiš, B., Štrok, M., 2011. Uptake of radionuclides by a common reed (Phragmites australis (Cav.) Trin. ex Steud.) grown in the vicinity of the former uranium mine at Žirovski vrh. Nucl. Eng. Des. 241, 1282–1286.
Chautard, C., Beaucaire, C., Gerard, M., Roy, R., Savoye, S., Descostes, M., 2020. Geochemical characterization of uranium mill tailings (Bois Noirs Limouzat, France) highlighting the U and ^{226}Ra retention. J. Environ. Radioact. 218, 106251.
Chen, S.B., Zhu, Y.G., Hu, Q.H., 2005. Soil to plant transfer of ^{238}U, ^{226}Ra and ^{232}Th on a uranium mining-impacted soil from southeastern China. J. Environ. Radioact. 82, 223–236.
Čučulović, A., Čučulović, R., Sabovljević, M., Radenković, M.B., Veselinović, D., 2016. Natural radionuclide uptake by mosses in eastern Serbia in 2008-2013. Arh. Hig. Rada. Toksiko. 67, 31–37.
da Conceição, F.T., Bonotto, D.M., Jiménez-Rueda, J.R., Roveda, J.A.F., 2009. Distribution of ^{226}Ra, ^{232}Th and ^{40}K in soils and sugar cane crops at Corumbataí river basin, São Paulo State, Brazil. Appl. Radiat. Isotopes 67, 1114–1120.
Didon-Lescot, J.-F., 1996. Forêt et développement durable au Mont-Lozère. Impact d'une plantation de résineux, de sa coupe et de son remplacement, sur l'eau et sur les réserves minérales du sol. (in French) PhD thesis, Université d'Orléans, Orléans, 210p.
Douay, F., Pruvot, C., Waterlot, C., Fritsch, C., Fourrier, H., Loriette, A., Bidar, G., Grand, C., de Vaufleury, A., Scheifler, R., 2009. Contamination of woody habitat soils around a former lead smelter in the North of France. Sci. Total Environ. 407, 5564–5577.
Dragović, S., Mihailović, N., Gajić, B., 2010. Quantification of transfer of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in mosses of a semi-natural ecosystem. J. Environ. Radioact. 101, 159–164.
Duchemin, B., Coursol, N., Bé, M.M., 1994. The re-evaluation of decay data for the U-238 chain. Nucl. Instrum. Methods A339, 146–150.
Durand, P., Neal, C., Lelong, F., Didon-Lescot, J.F., 1991. Hydrochemical variations in spruce, beech and grassland areas, Mont Lozere, southern France. J. Hydrol. 129, 57–70.
Ehlken, S., Kirchner, G., 2002. Environmental processes affecting plant root uptake of radioactive trace elements and variability of transfer factor data: a review. J. Environ. Radioact. 58, 97–112.
Georgiev, P., Groudev, S., Spasova, I., Nicolova, M., 2014. Ecotoxicological characteristic of a soil polluted by radioactive elements and heavy metals before and after its bioremediation. J. Geochem. Explor. 142, 122–129.
Gerzabek, M.H., Strebl, F., Temmel, B., 1998. Plant uptake of radionuclides in lysimeter experiments. Environ. Pollut. 99, 93–103.
Girault, F., Perrier, F., 2011. Heterogeneous temperature sensitivity of effective radium concentration from various rock and soil samples. Nat. Hazards Earth Syst. Sci. 11, 1619–1626.
Girault, F., Perrier, F., 2012a. Measuring effective radium concentration with large numbers of samples. Part I – experimental method and uncertainties. J. Environ. Radioact. 113, 177–188.
Girault, F., Perrier, F., 2012b. Measuring effective radium concentration with large numbers of samples. Part II – general properties and representativity. J. Environ. Radioact. 113, 189–202.
Girault, F., Perrier, F., 2019. Radon emanation from human hair. Sci. Total Environ. 660, 421–428.
Girault, F., Gajurel, A.P., Perrier, F., Upreti, B.N., Richon, P., 2011. Radon emanation of heterogeneous basin deposits in Kathmandu Valley, Nepal. J. Asian Earth Sci. 40, 595–610.
Girault, F., Perrier, F., Gajurel, A.P., Bhattarai, M., Koirala, B.P., Bollinger, L., Fort, M., France-Lanord, C., 2012. Effective radium concentration across the Main Central Thrust in the Nepal Himalayas. Geochim. Cosmochim. Acta 98, 203–227.
Girault, F., Perrier, F., Poitou, C., Isambert, A., Théveniaut, H., Laperche, V., Clozel-Leloup, B., Douay, F., 2016. Effective radium concentration in topsoils contaminated by lead and zinc smelters. Sci. Total Environ. 566-567, 865–876.
Girault, F., Perrier, F., Moreira, M., Zanda, B., Rochette, P., Teitler, Y., 2017a. Effective radium-226 concentration in meteorites. Geochim. Cosmochim. Acta 208, 198–219.
Girault, F., Schubnel, A., Pili, É., 2017b. Transient radon signals driven by fluid pressure pulse, micro-crack closure, and failure during granite deformation experiments. Earth Planet. Sci. Lett. 474, 409–418.
Girault, F., Perrier, F., Przylibski, T.A., 2018. Radon-222 and radium-226 occurrence in water: a review. Geol. Soc. Lond. S. P. 451, 131–154.
GIS Sol, 2011. L'état des sols de France. Groupement d'intérêt scientifique sur les sols. 188p. (in French).
Gunn, K.B., Mistry, K.B., 1970. The effect of chelating agents on the absorption of radium by plants. Plant Soil 33, 7–16.
Ham, G.J., Wilkins, B.T., Ewers, L.W., 2001. ^{210}Pb, ^{210}Po, ^{226}Ra, U and Th in arable crops and ovine liver: variations in concentrations in the United Kingdom and resultant doses. Radiat. Prot. Dosim. 93, 151–159.
Hassan, N.M., Ishikawa, T., Hosoda, M., Iwaoka, K., Sorimachi, A., Sahoo, S.K., Janik, M., Kranrod, C., Yonehara, H., Fukushi, M., Tokonami, S., 2011. The effect of water content on the radon emanation coefficient for some building materials used in Japan. Radiat. Meas. 46, 232–237.
He, Q., Walling, D.E., 1996. Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. J. Environ. Radioact. 30, 117–137.
Hu, N., Ding, D., Li, G., Zheng, J., Li, L., Zhao, W., Wang, Y., 2014. Vegetation composition and ^{226}Ra uptake by native plant species at a uranium mill tailings impoundment in South China. J. Environ. Radioact. 129, 100–106.
Ibrahim, S.A., Whicker, F.W., 1992. Comparative plant uptake and environmental behavior of U-series radionuclides at a uranium mine-mill. J. Radioanal. Nucl. Chem. 156, 253–267.
James, J.P., Dileep, B.N., Ravi, P.M., Joshi, R.M., Ajith, T.L., Hegde, A.G., Sarkar, P.K., 2011. Soil to leaf transfer factor for the radionuclides ^{226}Ra, ^{40}K, ^{137}Cs and ^{90}Sr at Kaiga region, India. J. Environ. Radioact. 102, 1070–1077.
Jha, V.N., Tripathi, R.M., Sethy, N.K., Sahoo, S.K., Shukla, A.K., Puranik, V.D., 2010. Bioaccumulation of ^{226}Ra by plants growing in fresh water ecosystem around the uranium industry at Jaduguda, India. J. Environ. Radioact. 101, 717–722.
Karunakara, N., Somashekarappa, H.M., Narayana, Y., Avadhani, D.N., Mahesh, H.M., Siddappa, K., 2003. ^{226}Ra, ^{40}K and ^{7}Be activity concentrations in plants in the environment of Kaiga, India. J. Environ. Radioact. 65, 255–266.
Kirchner, G., Daillant, O., 1998. Accumulation of ^{210}Pb, ^{226}Ra and radioactive cesium by fungi. Sci. Total Environ. 222, 63–70.
Krmar, M., Wattanavatee, K., Radnović, D., Slivka, J., Bhongsuwan, T., Frontasyeva, M.V., Pavlov, S.S., 2013. Airborne radionuclides in mosses collected at different latitudes. J. Environ. Radioact. 117, 45–48.
Lauria, D.C., Ribeiro, F.C.A., Conti, A.A., Loureiro, F.A., 2009. Radium and uranium levels in vegetables grown using different farming management systems. J. Environ. Radioact. 100, 176–183.
Li, Z., Ma, Z., van der Kuijp, T.J., Yuan, Z., Huang, L., 2014. A review of soil heavy metal pollution from mines in China: pollution and health risk assessment. Sci. Total Environ. 468–469, 843–853.
Lin, H., 2010. Earth's critical zone and hydropedology: concepts, characteristics, and advances. Hydrol. Earth Syst. Sci. 14, 25–45.
Luetzelschwab, J.W., Helweick, K.L., Hurst, K.A., 1989. Radon concentrations in five Pennsylvania soils. Health Phys. 56, 181–188.
Madruga, M.J., Brogueira, A., Alberto, G., Cardoso, F., 2001. ^{226}Ra bioavailability to plants at the Urgeiriça uranium mill tailings site. J. Environ. Radioact. 54, 175–188.

Manta, D.S., Angelone, M., Bellanca, A., Neria, R., Sprovieri, M., 2002. Heavy metals in urban soils: a case study from the city of Palermo (Sicily), Italy. Sci. Total Environ. 300, 229–243.
Markkanen, M., Arvela, H., 1992. Radon emanation from soils. Radiat. Prot. Dosim. 45, 269–272.
Markose, P.M., Bhat, I.S., Pillai, K.C., 1993. Some characteristics of ^{226}Ra transfer from soil and uranium mill tailings to plants. J. Environ. Radioact. 21, 131–142.
Marple, M.L., 1980. Ra-226 in Vegetation and Substrates at Inactive Uranium Mill Sites. Rep. LA-8183-T, Los Alamos Scientific Laboratory, Los Alamos, NM.
Martínez-Aguirre, A., Periáñez, R., 1998. Soil to plant transfer of ^{226}Ra in a marsh area: modelling application. J. Environ. Radioact. 39, 199–213.
Medley, P., Bollhöfer, A., 2016. Influence of group II metals on Radium-226 concentration ratios in the native green plum (*Buchanania obovata*) from the Alligator Rivers Region, Northern Territory, Australia. J. Environ. Radioact. 151, 551–557.
Medley, P., Bollhöfer, A., Parry, D., Martin, P., 2013. Radium concentration factors in passionfruit (*Passiflora foetida*) from the Alligator Rivers Region, Northern Territory, Australia. J. Environ. Radioact. 126, 137–146.
Moyen, C., Roblin, G., 2010. Uptake and translocation of strontium in hydroponically grown maize plants, and subsequent effects on tissue ion content, growth and chlorophyll a/b ratio: comparison with Ca effects. Environ. Exp. Bot. 68, 247–257.
Mrdakovic Popic, J., Oughton, D.H., Salbu, B., Skipperud, L., 2020. Transfer of naturally occurring radionuclides from soil to wild forest flora in an area with enhanced legacy and natural radioactivity in Norway. Environ. Sci.: Processes Impacts 22, 350.
Nathwani, J.S., Phillips, C.R., 1979. Adsorption of ^{226}Ra by soils in the presence of Ca^{2+} ions. Specific adsorption (II). Chemosphere 5, 293–299.
Nazaroff, W.W., 1992. Radon transport from soil to air. Rev. Geophys. 30, 137–160.
Nezami, S., Malakouti, M.J., Samani, A.B., Maragheh, M.G., 2016. Effect of low molecular weight organic acids on the uptake of ^{226}Ra by corn (*Zea mays* L.) in a region of high natural radioactivity in Ramsar-Iran. J. Environ. Radioact. 164, 145–150.
Nicolas, A., Girault, F., Schubnel, A., Pili, É., Passelègue, F., Fortin, J., Deldicque, D., 2014. Radon emanation from brittle fracturing in granites under upper crustal conditions. Geophys. Res. Lett. 41, 5436–5443.
Paul, A.C., Pillai, K.C., 1986. Transfer and uptake of radium in a natural and in a technologically modified radiation environment. J. Environ. Radioact. 3, 55–73.
Perrier, F., Aupiais, J., Girault, F., Przylibski, T.A., Bouquerel, H., 2016a. Optimized measurement of radium-226 concentration in liquid samples with radon-222 emanation. J. Environ. Radioact. 157, 52–59.
Perrier, F., Girault, F., Bouquerel, H., Bollinger, L., 2016b. Effective radium concentration in agricultural versus forest topsoils. J. Environ. Radioact. 160, 123–134.
Perrier, F., Girault, F., Bouquerel, H., 2018. Effective radium concentration in rocks, soils, plants, and bones. Geol. Soc. Lond. S. P. 451, 113–129.
Popovic, D., Todorovic, D., Frontasyeva, M., Ajtic, J., Tasic, M., Rajsic, S., 2008. Radionuclides and heavy metals in Borovac, Southern Serbia. Environ. Sci. Pollut. Res. 15, 509–520.
Przylibski, T.A., 2000. Estimating the radon emanation coefficient from crystalline rocks into groundwater. Appl. Radiat. Isotopes 53, 473–479.
Pulhani, V.A., Dafauti, S., Hedge, A.G., Sharma, R.M., Mishra, U.C., 2005. Uptake and distribution of natural radioactivity in wheat plants from soil. J. Environ. Radioact. 79, 331–346.
Righi, S., Bruzzi, L., 2006. Natural radioactivity and radon exhalation in building materials used in Italian dwellings. J. Environ. Radioact. 88, 158–170.
Rihs, S., Prunier, J., Thien, B., Lemarchand, D., Pierret, M.-C., Chabaux, F., 2011. Using short-lived nuclides of the U- and Th-series to probe the kinetics of colloid migration in forested soils. Geochim. Cosmochim. Acta 75, 7707–7724.
Ryan, B., Martin, P., Iles, M., 2005. Uranium-series radionuclides in native fruits and vegetables of northern Australia. J. Radioanal. Nucl. Chem. 264, 407–412.
Sakoda, A., Ishimori, Y., Yamaoka, K., 2011. A comprehensive review of radon emanation measurements for mineral, rock, soil, mill tailing and fly ash. Appl. Radiat. Isot. 69, 1422–1435.
Sam, A.K., Eriksson, Å., 1995. Radium-226 uptake by vegetation grown in Western Sudan. J. Environ. Radioact. 29, 27–38.
Sheppard, S.C., Evenden, W.G., 1988a. The assumption of linearity in soil and plant concentration ratios: an experimental evaluation. J. Environ. Radioact. 7, 221–247.
Sheppard, S.C., Evenden, W.G., 1988b. Critical compilation and review of plant/soil concentration ratios for uranium, thorium and lead. J. Environ. Radioact. 8, 255–285.
Sheppard, S.C., Sheppard, M.I., Tait, J.C., Sanipelli, B.L., 2006. Revision and meta-analysis of selected biosphere parameter values for chlorine, iodine, neptunium, radium, radon and uranium. J. Environ. Radioact. 89, 115–137.
Simon, S.L., Ibrahim, S.A., 1987. The plant/soil concentration ratio for calcium, radium, lead, and polonium: evidence for non-linearity with reference to substrate concentration. J. Environ. Radioact. 5, 123–142.
Simon, S.L., Ibrahim, S.A., 1990. Biological uptake of radium by terrestrial plants. In The Environmental Behaviour of Radium, Vol. 1, Technical Reports Series No. 310, IAEA, Vienna, 545-599.
Soudek, P., Petřík, P., Vágner, M., Tykva, R., Plojhar, V., Petrová, Š., Vaněk, T., 2007a. Botanical survey and screening of plant species which accumulate ^{226}Ra from contaminated soil of uranium waste depot. Eur. J. Soil Biol. 43, 251–261.
Soudek, P., Petrová, Š., Benešová, D., Tykva, R., Vaňková, R., Vaněk, T., 2007b. Comparison of ^{226}Ra nuclide from soil by three woody species *Betula pendula*, *Sambucus nigra* and *Alnus glutinosa* during the vegetation period. J. Environ. Radioact. 97, 76–82.
Soudek, P., Petrová, Š., Benešová, D., Kotyza, J., Vágner, M., Vaňková, R., Vaněk, T., 2010. Study of soil–plant transfer of ^{226}Ra under greenhouse conditions. J. Environ. Radioact. 101, 446–450.
Stoulos, S., Manolopoulou, M., Papastefanou, C., 2004. Measurement of radon emanation factor from granular samples: effects of additives in cement. Appl. Radiat. Isot. 60, 49–54.
Tanner, A.B., 1964. Radon migration in the ground: a review. In: Adams, J.A.S., Lowder, W.M. (Eds.), The Natural Radiation Environment. Univ. of Chicago Press, Chicago, pp. 161–190.
Thiry, Y., Van Hees, M., 2008. Evolution of pH, organic matter and ^{226}radium/calcium partitioning in U-mining debris following revegetation with pine trees. Sci. Total Environ. 393, 111–117.
Thu, H.N.P., Thang, N.V., Hao, L.C., 2020. The effects of some soil characteristics on radon emanation and diffusion. J. Environ. Radioact. 216, 106189.
Tuovinen, T.S., Kolehmainen, M., Roivainen, P., Kumlin, T., Makkonen, S., Holopainen, T., Juutilainen, J., 2016. Nonlinear transfer of elements from soil to plants: impact on radioecological modeling. Radiat. Environ. Biophys. 55, 393–400.
Uchida, S., Tagami, K., 2007. Soil-to-crop transfer factors of radium in Japanese agricultural fields. J. Nucl. Radiochem. Sci. 8, 137–142.
Uchida, S., Tagami, K., Shang, Z.R., Choi, Y.H., 2009. Uptake of radionuclides and stable elements from paddy soil to rice: a review. J. Environ. Radioact. 100, 739–745.
Uğur, A., Özden, B., Saç, M.M., Yener, G., 2003. Biomonitoring of ^{210}Po and ^{210}Pb using lichens and mosses around a uraniferous coal-fired power plant in western Turkey. Atmos. Environ. 37, 2237–2245.
Vandenhove, H., Van Hees, M., 2007. Predicting radium availability and uptake from soil properties. Chemosphere 69, 664–674.
Vandenhove, H., Eyckmans, T., Van Hees, M., 2005. Can barium and strontium be used as tracers for radium in soil–plant transfer studies? J. Environ. Radioact. 81, 255–267.
Vandenhove, H., Olyslaegers, G., Sanzharova, N., Shubina, O., Reed, E., Shang, Z., Velasco, H., 2009. Proposal for new best estimates of the soil-to-plant transfer factor of U, Th, Ra, Pb and Po. J. Environ. Radioact. 100, 721–732.
Vasconcellos, L.M.H., Amaral, E.C.S., Vianna, M.E., Penna Franca, E., 1987. Uptake of ^{226}Ra and ^{210}Pb by food crops cultivated in a region of high natural radioactivity in Brazil. J. Environ. Radioact. 5, 287–302.
Vera Tomé, F., Blanco Rodríguez, P., Lozano, J.C., 2002. Distribution and mobilization of U, Th and ^{226}Ra in the plant–soil compartments of a mineralized uranium area in southwest Spain. J. Environ. Radioact. 59, 41–60.
Vera Tomé, F., Blanco Rodríguez, M.P., Lozano, J.C., 2003. Soil-to-plant transfer factors for natural radionuclides and stable elements in a Mediterranean area. J. Environ. Radioact. 65, 161–175.
Vera Tomé, F., Blanco Rodríguez, P., Lozano, J.C., 2008. Elimination of natural uranium and ^{226}Ra from contaminated waters by rhizofiltration using *Helianthus annuus* L. Sci. Total Environ. 393, 351–357.
Vera Tomé, F., Blanco Rodríguez, P., Lozano, J.C., 2009. The ability of *Helianthus annuus* L. and *Brassica juncea* to uptake and translocate natural uranium and ^{226}Ra under different milieu conditions. Chemosphere 74, 293–300.
Wattanavatee, K., Krmar, M., Bhongsuwan, T., 2017. A survey of natural terrestrial and airborne radionuclides in moss samples from the peninsular Thailand. J. Environ. Radioact. 177, 113–127.
Weis, J.S., Weis, P., 2004. Metal uptake, transport and release by wetland plants: implications for phytoremediation and restoration. Environ. Int. 30, 685–700.
Whicker, F.W., Shaw, G., Voigt, G., Holm, E., 1999. Radioactive contamination: state of the science and its application to predictive models. Environ. Pollut. 100, 133–149.
Yan, X., Luo, X.G., 2016. Uptake of uranium, thorium, radium and potassium by four kinds of dominant plants grown in uranium mill tailing soils from the southern part of China. Radioprotection 51, 141–144.