Measuring effective radium concentration with large numbers of samples. Part II – general properties and representativity

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A R T I C L E   I N F O

Article history:
Received 9 January 2012
Received in revised form 16 May 2012
Accepted 18 June 2012
Available online 20 July 2012

Keywords:
Radon-222
Emanation
Radium
Temperature sensitivity
Moisture content
Building materials

A B S T R A C T

Effective radium concentration $EC_{Ra}$, product of radium concentration and radon emanation, is the source term for radon release into the pore space of rocks and the environment. Over a period of three years, we performed more than 6000 radon-222 accumulation experiments in the laboratory with scintillation flasks and SSNTDs and we obtained experimental $EC_{Ra}$ values from more than 1570 rock and soil samples. With this method, which allowed the measurement of $EC_{Ra}$ from large numbers of samples with sufficient accuracy and uncertainty, as detailed in the companion paper, the dependence of the emanation factor on temperature and moisture content is revisited. In addition, with such a large $EC_{Ra}$ dataset, dispersion of $EC_{Ra}$ can be studied at sample-scale (cm to dm) and at scarp-scale (m to tens of m). Furthermore, we are able to discuss the representativity of obtained $EC_{Ra}$ values at field-scale, and to investigate the spatial variations of $EC_{Ra}$ over kilometric scales, within geological formations and across formations and faults. This experimental study opens new perspectives in the understanding of radium geochemistry and illustrates the importance of studying the radon source term with large numbers of samples for the modelling of geological and environmental processes, and also for the assessment of the radon health hazard.

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1. Introduction

Radon-222, a radioactive noble gas with a half-life of about 3.8 days and daughter product of radium-226, accounts for half of the radiation dose to the general population (Porstendörfer, 1994) and is currently considered as a major source of lung cancer (Al-Zoughool and Krewski, 2009). The radon source term of natural samples is defined by the effective radium concentration $EC_{Ra}$, expressed in Bq kg$^{-1}$, which is the product of the radium concentration $C_{Ra}$ with the emanation coefficient $E$ (Stoulos et al., 2004). The emanation coefficient $E$ corresponds to the probability that a radium atom produces a radon atom which can reach the pore space of a soil or rock sample, and thus the atmosphere (Tanner, 1964; Nazaroff, 1992). While $C_{Ra}$ depends on the radium distribution in the sample, $E$, which varies from about 1% to more than 50% (Przylibski, 2000; Righi and Bruzzi, 2006) depends on the spatial distribution of radium in the sample and on the properties of the porous network. In addition, it can be affected by numerous external and internal factors (Sakoda et al., 2011). Among these factors, one can mention the water content of the sample (Barillon et al., 2005; Adler and Perrier, 2009) and temperature (Iskandar et al., 2004; Girault and Perrier, 2011). In order to understand a subtle physical property of rock or soil, such as $EC_{Ra}$, it is of utmost importance to carry out large numbers of $EC_{Ra}$ experiments from various rock and soil samples. However, few of the published papers dealing with the measurement of $EC_{Ra}$ from natural samples reported more than hundred $EC_{Ra}$ values.

During three years, as detailed in the companion paper (Girault and Perrier, 2012, JER, companion paper), we performed more than 6000 accumulations experiments to obtain $EC_{Ra}$ values from more than 1570 rock and soil samples. Accumulation experiments were conducted in the laboratory in air-tight containers (Stoulos et al., 2003) with both scintillation flasks and Solid State Nuclear Track Detectors (SSNTDs). The method appeared particularly useful, first, to obtain large numbers of $EC_{Ra}$ values and, second, to assess experimental uncertainties and detection limit using dedicated experiments including systematic checks and intercomparisons. As a result, experimental uncertainties were found sufficient for our purpose, ranging from 30% (1 σ) for $EC_{Ra}$ values smaller than 0.2 Bq kg$^{-1}$ to about 8—10% for $EC_{Ra}$ values larger than 50 Bq kg$^{-1}$. In addition, the detection limit, defined as the 90% probability for obtaining a non-zero experimental $EC_{Ra}$ value at 68% confidence level, varied between 0.04 and 0.09 Bq kg$^{-1}$. Thus, this experiment
was able to provide accurate \( E_{\text{Ra}} \) values from numerous types of samples encountered in the environment.

In this paper, we use the large \( E_{\text{Ra}} \) dataset obtained from more than 1570 rock and soil samples, whose uncertainties and detection limit are detailed in the companion paper (Girault and Perrier, 2012, JER, companion paper). To clarify the actual meaning of our \( E_{\text{Ra}} \) measurements, we study the importance of the preparation of the sample and discuss the representativity of the measurements of \( E_{\text{Ra}} \) in the field, hoping to pave the way to novel geological interpretations. While we regularly refer to previous papers, in particular for the detailed description of sites and geological context, all results and figures presented below are new.

2. Sensitivity of the measurement of \( E_{\text{Ra}} \) to experimental conditions

2.1. Preparation of sample

In most previous studies, samples were dried at about 105°C, but then there is no guarantee that the \( E_{\text{Ra}} \) measured under these conditions are relevant for field conditions. Not only the water content, known to affect the emanation coefficient, is an issue here, but the thermal stability of the water distribution, not necessarily possible to restore by wetting, but also chemical reactions may occur, especially for soil samples. In addition, biofilms, destroyed by heat, might play an important role in radon emanation. Such issues have not been studied yet, but might be accessible with accurate measurements using large samples. In our studies, except when stated otherwise, we do not make any thermal treatment on the samples.

For rock and soil samples, an important issue is the spatial distribution of radium, which can be addressed in part by studying \( E_{\text{Ra}} \) versus sample grain size (Sakoda et al., 2010; Breitner et al., 2010). For this purpose, the experimental uncertainties must be adequate enough, especially at low values of \( E_{\text{Ra}} \). This is possible using our methodology, as illustrated in Fig. 1a. In this figure, the ratio of \( E_{\text{Ra}} \) for grain size smaller than 100 μm to the \( E_{\text{Ra}} \) for grain size larger than 100 μm is shown versus \( E_{\text{Ra}} \) for 14 crushed gneiss samples from the Roselend dead-end tunnel in the French Alps (Richon et al., 2005). The two different grain sizes are separated by sieving after machine crushing. Two groups of samples emerge from Fig. 1a. For 9 samples, the values of the ratio cluster around 2, indicating a definite enhancement of \( E_{\text{Ra}} \) for the finer phase, which might be due to either an enhancement of radium concentration, or an enhancement of \( E \), or both. This enhancement of radon release from the finer phases has been observed in numerous cases (Markkanen and Arvela, 1992; De Martino et al., 1998; Garver and Baskaran, 2004; Tuccimei et al., 2006; Somlai et al., 2008; Breitner et al., 2010), and attributed to preferential adsorption of radium on clay minerals (Ames et al., 1983a, 1983b; Hidaka et al., 2007). In Fig. 1a, however, five samples show ratios compatible with 1, thus showing no enhancement. This enhancement, thus, is not a general fact, even within a given geological formation and similar sampling conditions. It may however indicate subtle differences in local bedrock or weathering conditions, which might deserve further studies.

Another example of grain size effect is depicted in Fig. 1b, for two rock samples, Bajocian schist and Houiller sandstone, from Sur- Frêtes and La Gittaz in the French Alps (Trique et al., 2002). Finer grain size appears to enhance \( E_{\text{Ra}} \) values, in particular for the sandstone sample, by doubling the initial value. Results are more contrasted for the schist sample, which show similar \( E_{\text{Ra}} \) values for both the finer and the larger grain size fractions (Fig. 1b). These observations may imply important effect on \( E \) of the radium distribution in particles of different size (Cosma et al., 2001; Girault and Perrier, 2012, JER, companion paper).

For a rock sample, one most important issue is the comparison between the total effective radium concentration \( TEC_{\text{Ra}} \) which is measured by an accumulation experiment in which radon diffusion effects are negligible (crushed sample) and the apparent effective radium concentration \( AEC_{\text{Ra}} \) which corresponds to only some fraction of the \( TEC_{\text{Ra}} \) that are measurable when diffusion effects occur (bulk rock sample). Examples of such comparisons are shown in Fig. 2, which displays the normalized difference:

\[
D_i = \frac{E_{\text{Ra}}^{\text{crushed}} - E_{\text{Ra}}^{\text{rock}}}{\sqrt{\sigma_i^{\text{crushed}} + \sigma_i^{\text{rock}}}}
\]

for selected samples (diamonds) for which the \( E_{\text{Ra}} \) was first measured with a rock sample, then remeasured in the same conditions.
conditions after crushing. The loss of finer phases during crushing was avoided as much as possible by careful manual crushing, in order to avoid the possible bias in $EC_{Ra}$ described before.

The average value of the distribution is: $0.69 \pm 0.45$, thus showing no statistically significant evidence for reduction of $EC_{Ra}$ for bulk rock samples, which means that the radon diffusion length in these rocks is larger than the typical rock size used in the experiments, namely a few centimetres. Standard deviation is $3.43 \pm 0.48$, thus suggesting some dispersion effects beyond the experimental uncertainties. The same conclusions are drawn when $EC_{Ra}$ of a bulk subsample is compared with the $EC_{Ra}$ of another crushed subsample (triangles in Fig. 2), again showing no systematic significant reduction of the average ($-0.30 \pm 0.53$). The samples used in these tests are all granites and gneiss samples, from various locations including the Roselend tunnel in the French Alps, and from crystalline metamorphic units in Nepal. These rocks, affected by Miocene and recent tectonics, probably contain significant microfracturation, which then tends to increase the effective diffusion coefficient of radon. While we found no reduction of $EC_{Ra}$ in the tested rock samples, our study shows that the experimental methodology is sufficiently accurate to be able to observe the phenomenon, if present.

This test shows that, for routine measurements, it is better to crush the samples, even in a coarse manner, so that the samples can be measured using bottles with narrow throats, which gave the best reproducibility and the smaller amount of leakage problems during our study (Girault and Perrier, 2012, JER, companion paper).

2.2. Effective radium concentration versus temperature

Temperature is known to increase the emanation coefficient and, thus, the $EC_{Ra}$ value in a given sample (Iskandar et al., 2004; Lee et al., 2010). This effect, however, is small and, to observe it in a reliable manner, high precision experiments are needed. Our accumulation experiments with scintillation asks have already revealed their usefulness in different rock and soil samples (Girault and Perrier, 2011). Indeed, repeated measurements carried out at three distinct temperatures (7, 22 and 37 °C) provide a sufficiently precise estimation of the enhancement of $EC_{Ra}$ with temperature. Temperature sensitivity (TS) from rock and soil samples, defined as the slope of the fit of the experimental value of $EC_{Ra}$ normalized by $EC_{Ra}$ at 20 °C, ranged from 0.16 to 2.0% °C$^{-1}$ and from 0.10 to 2.0% °C$^{-1}$, respectively (Girault and Perrier, 2011).

In the present study, we selected two rock and two soil samples from this previous dedicated experiment in order to refine their TS. Two new $EC_{Ra}$ measurements at two additional temperature stages (32 and 46 °C) have thus been performed. These additional results are shown in Fig. 3 together with the previous results for the four samples. The increasing trend of $EC_{Ra}$ is roughly linear for the entire sample set. At 32 °C, generally, $EC_{Ra}$ values are observed lower than the linear enhancement. Moreover, while the increase of $EC_{Ra}$ at temperature larger than 40 °C may be diminished, the linear approximation remains valid. For rock samples, we obtained TS values of $0.75 \pm 0.20$ and $1.09 \pm 0.14$ °C$^{-1}$ for R3 and R8, respectively, whereas TS from soil samples are lower, 0.58 ± 0.09 and 0.61 ± 0.07 °C$^{-1}$ for S1 and SN2, respectively. The error bars on TS are calculated by randomly removing one point from the data set. For two samples, R3 and SN2, the TS value determined using 5 temperature stages is compatible with the former study, in which 3 temperature stages only were considered. Indeed, differences are of the order of 20% or smaller (2.7 ± 1.0% for R3). However, for R8 and S1, differences are larger than 50%, confirming undoubtedly the need of repeated measurements for reliable TS estimation.

At this stage, it may not be easy to explain theoretically the observed TS of $EC_{Ra}$ in a conclusive manner. Nevertheless, in a first approximation, one can investigate the importance of indirect effect of water content and adsorption. Indeed, what is actually measured in the accumulation experiments is the apparent effective radium concentration, which is related to the true effective radium concentration by Meslin et al. (2011):

$$EC_{Ra}^{app} = \frac{\varepsilon_a + \varepsilon_w}{\varepsilon_a + \varepsilon_w K_w + \rho a} EC_{Ra}$$

where $\varepsilon_a$ is the air connected porosity, $\varepsilon_w$ the water connected porosity, $K_w$ the dimensionless radon water/air partition coefficient (Henry’s constant), $\rho$ the radon adsorption coefficient (m$^2$ kg$^{-1}$) and $a$ the density of the sample (kg m$^{-3}$). From Eq. (2), TS can result from temperature sensitivity of $K_w$ or temperature sensitivity of $\varepsilon_w$.

The variation of $\varepsilon_w$ with temperature is well known (Weigel, 1978). Here, we use an updated parameterization:

$$K_w(T_C) = 0.104 + 0.416e^{-0.0491T_C},$$

where $T_C$ is the temperature in °C. The physics of radon adsorption is less well known (Sceryh and Whittlestone, 1989). In the following, we use the expression as compiled by Meslin et al. (2011):

$$K_w(T) = K_0^w e^{-135e^{-6T}},$$

where $K_0^w$ equals to $5.5 \times 10^{-4} m^2 kg^{-1}$. $Q_w$ is the adsorption heat ($2 \times 10^4$ J mol$^{-1}$). $R$ is the ideal gas constant and $T$ is the temperature expressed in K.

In Fig. 4, for three sets of parameters, effects of temperature dependence of $\varepsilon_w$ and $K_w$ are presented as dashed and full lines, respectively, for the ratio $EC_{Ra}^{app}/EC_{Ra}$ (20 °C) and the TS. With the given choice of parameters, the observed range of TS, from 0.1 to 3% °C$^{-1}$, can be easily covered. The effect of the temperature dependence of $K_w$ on TS of a given sample is larger than the effect from the temperature dependence of $\varepsilon_w$. Moreover, intermediate water content implies low TS values, of the same order of magnitude than the effect of $\varepsilon_w$. At constant water content, smaller porosity values for the sample result in larger TS. This enhancement for small porosity might account for the large TS of about 3% °C$^{-1}$ reported for pitchblende grains (Perrier and Girault, 2012). Therefore, temperature dependence of the adsorption coefficient, in general, seems sufficient to accommodate the observed TS. Adsorption effects may not be the only factors explaining the heterogeneity of the TS of $EC_{Ra}$ but it definitely appears as a reasonable candidate.

2.3. Effective radium concentration versus water content

The water content is known to affect the emanation coefficient of radon (Sakoda et al., 2011). Residual water plays an important

![Fig. 3. $EC_{Ra}$ versus temperature for two soil samples (S1 and SN2) and two rock samples (R3 and R8). $EC_{Ra}$ values obtained at 32 °C and 46 °C are added to previous data from Girault and Perrier (2011).](image-url)
role in the pore space in slowing down the recoil radon atoms from radium decay (Tanner, 1964), an effect which is now reasonably well understood in porous media (Semkow, 1991; Barillon et al., 2005; Adler and Perrier, 2009). When pores are saturated with water, by contrast, apparent radon exhalation flux from the sample is reduced because of a dramatic reduction of the effective diffusion coefficient (Meslin et al., 2010). This behaviour is confirmed by experimental measurements (Fleischer, 1987; Sun and Furbish, 1995; Bossew, 2003; Breitner et al., 2010; Sakoda et al., 2010; Hassan et al., 2011). However, the number of available experimental data remains scarce.

While the effect is rather small, it is observable given our experimental accuracy. For example, for the SBC1R2 soil (see the companion paper) we carried out three measurements using three subsamples. For the subsample in natural conditions, without treatment, we obtain $EC_{Ra} = 0.3 \pm 0.1$ Bq kg$^{-1}$. The relative massic moisture content of this sample, measured after the accumulation experiments, is $x_w = 12\%$. For one subsample in which water has been added by spraying, we obtain $EC_{Ra} = 27.2 \pm 1.3$ Bq kg$^{-1}$, for $x_w = 27\%$. For one subsample left drying in a metal pan exposed to the sun during 5 h, we obtain $EC_{Ra} = 25.5 \pm 1.0$ Bq kg$^{-1}$. The intermediate water content observed in natural conditions thus leads to an enhanced $EC_{Ra}$ with an enhancement of about 10–20%.

Detailed experiments were conducted to measure more accurately, with the same sample in the same conditions, the effect of moisture on the emanation coefficient. The results from these experiments, performed with 6 rock samples and 6 soil samples, are collected in Fig. 5. To avoid interference with the temperature sensitivity of $EC_{Ra}$, which is also dependent on the moisture content (Breitner et al., 2010), these accumulation experiments were conducted in a temperature controlled environment, with a temperature set at 25°C, with temporal variations smaller than 0.1 °C. Temperature during the experiments was monitored with Tintag™ Talk temperature monitors (Gemini Data Loggers, UK) with a sampling time of 30 min. The samples were dried in desicators instead of heating.

To first order, the variation of $EC_{Ra}$ with respect to water content, expressed in mass per cent of the dry sample, is similar for the six rock samples in Fig. 5a and b. Indeed, a sharp $EC_{Ra}$ decrease is observed from 1–5% to 0% of water, with a global average factor of 1.6 ± 0.2, sometimes reaching more than 2 (R4 and R6). At larger water content, $EC_{Ra}$ decreases progressively from 5 to 10% of water and, for three samples (R3, R5 and R6), the reduction is more important, sometimes larger than the value obtained for zero water content (R3 sample). For all rock samples studied, $EC_{Ra}$ appears relatively stable between 2 and 10% of water.

Looking in more details, while the observed behaviour is similar among the rock samples, more differences are seen among the soil samples (Fig. 5c). Indeed, $EC_{Ra}$ diminishes strongly from 2–3% to 0% of water, with an average factor of 1.8 ± 0.2, with a maximum of 2.3 ± 0.2 for S5 sample. Some samples do not show any reduction of $EC_{Ra}$ at low water content. This may be due to the fact that all soil samples were still not entirely dried. Nonetheless, for four samples (S1–4), the reduction of $EC_{Ra}$ is also important at large water content, typically from 20 to 25% only. In addition, $EC_{Ra}$ is stable over a larger range of water content than for rock samples, from 5 to 20% in general, but sometimes up to 30–35% (S1 and S2).

The range of variation of $EC_{Ra}$ with water content is compatible both for rock and soil samples with reported variation of the radon emanation coefficient $E$. Indeed, for both samples, $E$ increases from 0% to some % of water by a factor of 2, with a maximum between 5 and 15% (Menetrez et al., 1996; Barillon et al., 2005; Adler and Perrier, 2009). In our experiments, maximum is reached between 1 and 14% and between 2 and 25% for rock and soil samples, respectively. The sharp decrease observed at low water content can be larger than a factor 2 (Markkanen and Arvela, 1992), with a maximum of about 6 for a Martian regolith (Meslin et al., 2011).

In order to evaluate the importance of the effect of water content in practical situations, it is useful to have at hand some simplified parameterization. For this purpose, we propose an analytical expression using two exponential terms, depending on the volumetric water saturation $S_{w}$. Therefore, we scale the previous results as a function of the estimated volumetric water content.

![Fig. 4. Effect of the temperature dependence of radon water/air partition coefficient $x_w$ (dashed lines) and radon adsorption coefficient $k_d$ (full lines) on the temperature sensitivity $TS$ of $EC_{Ra}$. For three sets of parameters (adsorption $k_d$, porosity $\epsilon$ and water content $S_w$), the top graph shows the $EC_{Ra}/EC_{Ra}(20\degree C)$ ratio versus temperature, while the bottom graph shows $TS$ versus temperature.](image)

![Fig. 5. $EC_{Ra}$ versus water content in mass per cent of the dry sample for (a, b) six crushed rock samples (R1–6) and (c) six soil samples (S1–6).](image)
3. Representativity of $EC_{Ra}$

Sample-scale dispersion (SASD) is the standard deviation of $EC_{Ra}$ observed over a set of subsamples of the same original sample. This concept applies for rock samples, but also for soil samples. Indeed, a soil sample of about 100–200 g is not necessarily a fully mixed system, but may contain organized clusters and lumps. Thus, the effect of water saturation will not be a concern in soils, whereas it remains an important factor for rocks. For example, in the case of the SYP12 Compreignac granite drill core, subsamples, separated by less than a few cm, showed relatively large variations in the values of $EC_{Ra}$, varying between $329 \pm 11$ and $719 \pm 25$ Bq kg$^{-1}$, with an average of $556 \pm 39$ Bq kg$^{-1}$ and SASD of 22 ± 5%. For the Syabru-Bensi augen gneiss, one sample was collected and subdivided into 10 subsamples. These subsamples showed moderate variations in the $EC_{Ra}$ values, with an average of $241 \pm 9$ Bq kg$^{-1}$ and SASD of 13 ± 3%. Other granite subsamples collected in France gave moderate SASD of 9–16%.

This dispersion, varying from 4 to 22% is not necessarily easy to explain. Differences must be due to heterogeneity of mineralization, in particular for secondary minerals, redepension after

### Table 1

Examples of sample-scale dispersion (SASD) of $EC_{Ra}$ obtained by studying the $EC_{Ra}$ of a number of subsamples from a given sample.

<table>
<thead>
<tr>
<th>Sample Description</th>
<th>Number of subsamples</th>
<th>Average (Bq kg$^{-1}$)</th>
<th>Min/Max (Bq kg$^{-1}$)</th>
<th>SASD (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soils Syabru-Bensi gas discharge zone SBC1R2 (Nepal)</td>
<td>15</td>
<td>30.7 ± 0.9</td>
<td>25.5 ± 1.0/37.2 ± 4.0</td>
<td>12 ± 2</td>
</tr>
<tr>
<td>Vaugirgneuse village near Paris (France)</td>
<td>12</td>
<td>5.70 ± 0.07</td>
<td>5.28 ± 0.19/6.04 ± 0.21</td>
<td>4.2 ± 0.9</td>
</tr>
<tr>
<td>Soil overlying Anduze granite (France)</td>
<td>12</td>
<td>26.8 ± 0.5</td>
<td>22.8 ± 0.5/29.1 ± 0.5</td>
<td>7 ± 1</td>
</tr>
<tr>
<td>Rocks Compreignac granite SYP12 (France)</td>
<td>10</td>
<td>556 ± 39</td>
<td>329 ± 11/719 ± 25</td>
<td>22 ± 5</td>
</tr>
<tr>
<td>Syabru-Bensi augen gneiss SYAS (Nepal)</td>
<td>11</td>
<td>240.8 ± 9.3</td>
<td>201 ± 4/286 ± 4</td>
<td>13 ± 3</td>
</tr>
<tr>
<td>Anduze granite AND485 (France)</td>
<td>2</td>
<td>52.9 ± 2.4</td>
<td>49.5 ± 1.8/56.4 ± 4.5</td>
<td>9 ± 5</td>
</tr>
<tr>
<td>St-Sylvestre granite BESS (France)</td>
<td>5</td>
<td>74.0 ± 5.2</td>
<td>64.8 ± 9.4/94.2 ± 10.5</td>
<td>16 ± 5</td>
</tr>
</tbody>
</table>
leaching or weathering (Sakoda et al., 2008). Indeed, all rock samples are located in weathered outcrops. This argument however does not apply to the Compreignac granite drill core. Indeed, this core was confined at depth and thus protected from the strongest weathering processes. Heterogeneities are likely due to rich-radium grains, oxides and minerals which were not homogeneously distributed initially. The SASD thus addresses fundamental questions relative to the nature and context of the considered sample.

3.2. Stratigraphic dispersion and coherence for soils

The $E_{\text{Ra}}$ values from soil samples in the Kathmandu valley (Girault et al., 2011a) vary from 0.4 to 43 Bq kg$^{-1}$, depending on the soil characteristics (sand, mud or clay) and have a remarkable consistency in a given horizon. Therefore, such technique can be used for stratigraphic correlation at long distance, from metres to kilometres. For different sedimentary horizons, several samples were collected in a given horizon. From the published results of this study, dispersion of $E_{\text{Ra}}$ values in a considered horizon is presented in Table 2. In addition, thirteen top soils have been sampled near Paris and near Anduze (Cévennes).

Examples of scarp-scale dispersion (SCSD) were taken from the two sites described before, namely near Paris and near Anduze (Cévennes).

For soil samples from the Kathmandu valley, the scarp–scale dispersion (SCSD) was relatively small, similar to the observed SASD. The SCSD of 27.3% and 6.8% for orange-greenish clay to 22 ± 8% for black clay (Table 2), with a consistent overall average of 18 ± 1%. Furthermore, in three entire scarp series comprising sand and clay of various horizons, despite the fact that they were separated by 1 km and by 2.5 km, $E_{\text{Ra}}$ values were compatible and showed low SCSD of 27.3 ± 6.8% (Girault et al., 2011a). Soils from the leucogranite area in central Nepal show similar SCSD of 22 ± 4%. For the soil samples from France, the SCSD of Vaugrigneuse soils follows the trend indicated by the SASD; it was lower (12 ± 2%) than the SCSD of Anduze soils (25 ± 5%).

The consistency for soils appears thus remarkable even among distant scarps. The stability of $E_{\text{Ra}}$ values in a given horizon may reflect the relatively homogeneous deposition process and, in a less significant manner, homogeneity of the weathering process at a given time in a given sedimentary horizon. This point reveals a potential usefulness for $E_{\text{Ra}}$ studies in stratigraphy and long distance correlations of soil scarps.

3.3. Scarp-scale dispersion for rocks and scarp consistency

To be able to study the relationship between $E_{\text{Ra}}$ and geology, the stability of the $E_{\text{Ra}}$ values at scarp level must be studied. Examples are given below.

In the Roselend dead-end tunnel, the gneiss formation was sampled at the roof surface every 10 m in the 124-m-long tunnel (Fig. 7a), and in a drill core PERM4 (Wassermann et al., 2011) located in the end part of the tunnel (Fig. 7b), for which $E_{\text{Ra}}$ values are presented as a function of distance from the tunnel in Fig. 7c. In Fig. 7a, the enhancement of $E_{\text{Ra}}$ for the <100 µm crushed phase compared with the >100 µm crushed phase, discussed previously, can also be seen at some locations in the tunnel. The similarity between $E_{\text{Ra}}$ of bulk and crushed rock samples is also visible in this figure. While the tunnel shows some geological heterogeneity in

Table 2 Examples of scarp-scale dispersion (SCSD) of $E_{\text{Ra}}$, obtained by studying the $E_{\text{Ra}}$ of a number of rock or soil samples taken at distances of the order of a few metres maximum from each other, in the same geological scarp or in the same soil formation.

<table>
<thead>
<tr>
<th>Transect</th>
<th>Number of samples</th>
<th>Average (Bq kg$^{-1}$)</th>
<th>Min–Max (Bq kg$^{-1}$)</th>
<th>SCSD (%)</th>
</tr>
</thead>
</table>
| Soils
| Kathmandu Valley (Nepal)
| (figures in Girault et al. (2011a))
| Massive Black Mud (Fig. 4) | 13 | 10.89 ± 0.45 | 7.7–13 | 15 ± 3 |
| Black clay (Fig. 4) | 10 | 25.1 ± 1.5 | 20–34 | 19 ± 4 |
| Top soil (Fig. 9) | 8 | 9.63 ± 0.66 | 7.0–13 | 19 ± 5 |
| Orange-greenish clay (Fig. 9) | 4 | 26.1 ± 1.6 | 23–31 | 13 ± 4 |
| Top soil (Fig. 10) | 4 | 3.56 ± 0.38 | 2.9–4.6 | 21 ± 8 |
| Orange-greenish clay (Fig. 10) | 4 | 28.1 ± 1.6 | 24–31 | 11 ± 4 |
| Black clay (Fig. 10) | 4 | 30.2 ± 3.3 | 23–37 | 22 ± 8 |
| Red residual soil (Fig. 11) | 10 | 10.00 ± 0.69 | 6.3–14 | 22 ± 5 |
| Central Nepal (Langtang): Kyanjin Gompa top soil | 13 | 14.4 ± 2.1 | 24–53 | 22 ± 4 |
| Vaugrigneuse village near Paris (France) top soil | 12 | 5.25 ± 0.18 | 4.4–6.4 | 12 ± 2 |
| Soil overlying Anduze granite (France) | 12 | 32.3 ± 2.4 | 24–51 | 25 ± 5 |
| Rocks
| Roselend tunnel gneiss (France)
| Along tunnel (Fig. 7a) | 20 | 2.52 ± 0.25 | 1.0–4.8 | 44 ± 8 |
| PERM4 (Fig. 7c) | 12 | 1.13 ± 0.17 | 0.55–2.4 | 53 ± 12 |
| All Roselend gneiss samples | 32 | 2.0 ± 0.2 | 0.55–4.8 | 58 ± 8 |
| Vincennes Lutetian limestone (France)
| Top bancs francs | 13 | 1.44 ± 0.19 | 0.66–3.2 | 48 ± 10 |
| Building limestone | 6 | 0.27 ± 0.04 | 0.15–0.38 | 32 ± 10 |
| Clay layers | 2 | 4.06 ± 0.41 | 3.7–4.5 | 14 ± 7 |
| All Vincennes quarry samples (Fig. 10) | 22 | 1.39 ± 0.24 | 0.15–4.5 | 82 ± 16 |
| Lesser Himalaya rocks (Nepal)
| Black slate (Fig. 9) | 13 | 44.7 ± 4.8 | 20–73 | 38 ± 8 |
| Calc-schist FA (Fig. 8) | 12 | 1.72 ± 0.23 | 0.62–3.4 | 46 ± 10 |
| Calc-schist and mica-schist FB (Fig. 8) | 12 | 2.63 ± 0.47 | 0.66–5.3 | 61 ± 15 |
| FB Fresh samples (Fig. 8) | 7 | 1.41 ± 0.17 | 0.66–2.0 | 32 ± 9 |
| FB Weathered samples (Fig. 8) | 5 | 4.35 ± 0.34 | 3.6–5.3 | 17 ± 6 |
| Syabru-Bensi Augen gneiss | 12 | 14.8 ± 1.7 | 3.9–26 | 40 ± 9 |
| Ulleri Augen gneiss | 11 | 19.5 ± 1.9 | 8.8–28 | 32 ± 7 |
| Anduze granite (France)
| Reference transect | 12 | 27.8 ± 2.4 | 19–43 | 29 ± 6 |
| Transect 50 cm lower | 12 | 25.7 ± 2.0 | 16–40 | 27 ± 6 |
the gneiss (Pili et al., 2004), with pegmatite bodies conspicuous within the gneiss in the end part of the tunnel, the obtained EC$_{\text{Ra}}$ values in the Roselend tunnel were remarkably consistent, with a rather low average value of 2.52 ± 0.25 Bq kg$^{-1}$, roughly consistent with one result 1.1 ± 0.3 Bq kg$^{-1}$ obtained with an Alphaguard continuous radon monitor (Richon et al., 2005). Overall SCSD was rather moderate, with a value of 58 ± 8% (Table 2). Some spatial structure is nevertheless observed along the tunnel, with a smooth increase from the entrance towards the middle section and the inner room, and a gradual increase towards the end of the tunnel. This spatial variation of EC$_{\text{Ra}}$ is compatible with the spatial variation of the gamma dose rate measured along the tunnel (Richon et al., 2005).

The smoothness of the spatial variation is especially clear along the PERM4 borehole, which showed a remarkable spatial consistency at spatial scales of the order of 20 cm, better than along the tunnel. Some enhancements of EC$_{\text{Ra}}$ were also observed after 1 m in the borehole, with an isolated higher value at 1.5 m distance and a more continuous enhancement deeper in the bedrock (Fig. 7c). The measurement of EC$_{\text{Ra}}$ appears as a useful tool when mapping the bedrock properties and this study suggests that more ambitious campaigns should be attempted. While the number of data remains insufficient, the results from Fig. 7 suggest a layered structure around the tunnel, which might reflect the extend of the Excavation Damage Zone (EDZ), but could also reflect the overall heterogeneity of the gneiss in the tunnel, as indicated by Fig. 7a. These samples, included in Fig. 2, did not show any significant enhancement of EC$_{\text{Ra}}$ for the crushed drill cores compared with the bulk drill cores, suggesting that radon diffusion length, along the whole PERM4 profile, is consistently larger than a few cm. This is even the case for the end sample, at 2.5 m distance, for which one may have anticipated less microfracturation outside of the potential EDZ (Wassermann et al., 2011), and for which we have $AEC_{\text{Ra}} = 2.44 ± 0.13$ Bq kg$^{-1}$ and $TEC_{\text{Ra}} = 2.23 ± 0.13$ Bq kg$^{-1}$. Another possibility, which cannot be ruled out, is that the radius of the EDZ at the PERM4 location is in fact much larger than the borehole depth.

In the Lesser Himalayan Sequences, near Syabru-Bensi (Central Nepal), two outcrops were studied in details. The FA outcrop is comprised of calc-schist rocks (Fig. 8a and b). Twelve samples were collected at this outcrop along two parallel profiles, one above the other. The EC$_{\text{Ra}}$ values both showed consistency along a given profile and compatibility within the whole sample set at this site. The global average was 1.7 ± 0.2 Bq kg$^{-1}$, with a SCSD of 46 ± 10% (Table 2). Larger values were observed, in some cases resulting in a doubling of EC$_{\text{Ra}}$ values (sample 2 in Fig. 8b). This sample 2 may be more mica-rich, leading to an enhancement of initial radium concentration by adsorption. The second outcrop considered here (FB) consists of a contact between calc-schist and mica-schist, which is in fact difficult to place with sufficient accuracy (Fig. 8c and d). Along this outcrop, rocks were characterized by similar EC$_{\text{Ra}}$ values, to first order, thus signifying that the contact between the two different rocks could be some metres further north. The global average was 2.6 ± 0.5 Bq kg$^{-1}$, with large SCSD of 61 ± 15% (Table 2). Seven samples showed ochre secondary deposition, which may more likely result from garnet alteration by precipitation and leaching. Other samples did not show any trace of weathering of that type. Along this outcrop, weathered samples had larger EC$_{\text{Ra}}$ values than unweathered samples, with averages 4.4 ± 0.3 and 1.4 ± 0.2 Bq kg$^{-1}$, respectively. Their respective SCSD is 17 ± 6% and 32 ± 9% (Table 2). This example is particularly interesting and implies that it is of utmost importance to avoid sampling rocks in strong weathering locations, in particular when workers are willing to collect only a few samples to characterize the entire outcrop or the whole formation.

In the north-western part of the Kathmandu valley (Central Nepal), one black slate outcrop belonging to the Lesser Himalayan

Fig. 7. EC$_{\text{Ra}}$ of gneiss samples from the Roselend tunnel in the French Alps (Richon et al., 2005; Wassermann et al., 2011) versus position in the tunnel (a), whose layout is shown in (b). Most samples were crushed and the EC$_{\text{Ra}}$ values are shown separately for grain size smaller and larger than 100 μm. Three EC$_{\text{Ra}}$ values (grey filled symbols) are also available for uncrushed rock pieces and crushed aliquots of the same sample. In inset (c), EC$_{\text{Ra}}$ values of drill cores from the PERM4 borehole, located in (b), are shown versus their position from the tunnel.
Sequences have been sampled. A total of 6 samples were collected horizontally at large distance (more than 40 m), whereas the 7 other samples were collected horizontally at small distance, with one sample separated by 40 cm maximum from the next one. Samples location and $E_{Ra}$ results are depicted in Fig. 9. For the whole sample set, $E_{Ra}$ values showed an average of $45 \pm 5 \text{ Bq kg}^{-1}$ with a range of variation from 20 to 73 Bq kg$^{-1}$. The SCSD was relatively large: $38 \pm 8\%$ (Table 2). The most interesting point lies in the observation that, whatever the observable spatial scale, $E_{Ra}$ values showed similar dispersion (Fig. 9b). In such a case, thus, it is not necessary to collect samples at only large or only small distance along a given outcrop to obtain a significant $E_{Ra}$ average value.

Other outcrops were studied north of Syabru-Bensi (Central Nepal) and near Ulleri village (Mid-Western Nepal). Both consist of augen gneisses, characterized by granitic lenses. At each outcrop, a dozen samples were collected and their results are summarized in Table 2. These two gneiss occurrences belong to the Ulleri augen gneiss formation, which is reported within the Lesser Himalayan Sequences throughout the Nepalese Himalayas. At Syabru-Bensi location, the $E_{Ra}$ average value was $15 \pm 1.7 \text{ Bq kg}^{-1}$, with SCSD of $40 \pm 9\%$. At Ulleri locality, the $E_{Ra}$ average value was $20 \pm 1.9 \text{ Bq kg}^{-1}$, with SCSD of $32 \pm 7\%$. Although they were separated by about 160 km, the characteristics of $E_{Ra}$ appear noteworthy similar, even when taking into account their respective range of variation (Table 2). Consequently, this example demonstrates the potentiality of $E_{Ra}$ at large spatial scales. Another example of impressive interest for large spatial scale $E_{Ra}$ studies is detailed in the following section.

In the south of France, two horizontal transects of 12 rock samples separated vertically by 50 cm were taken from an Anduze granite outcrop. The distance between two samples of the same transect is 50 cm. When comparing the two transects as a whole, all characteristics of the $E_{Ra}$ values are consistent. Indeed, for each transects, the average, the range of variation as well as the SCSD appears similar. The obtained SCSD is low, from 27 to 29%, but larger than the average SCSD of soils of our study (Table 2).

Compared with the three previous situations, spatial variations of $E_{Ra}$ can be rather different in the context of sedimentary rocks, as illustrated in Fig. 10, which shows $E_{Ra}$ values measured over a scarp of Lutetian limestone in the Vincennes underground quarry, in the Paris Basin (Perrier and Richon, 2010). While the average value remained low, $1.39 \pm 0.24 \text{ Bq kg}^{-1}$, but comparable with the average value observed in the Roselend gneiss (see Table 2), large spatial variations were observed along the vertical direction perpendicular to the stratification. The thick and in general highly porous limestone beds, located below the Souchet interface, showed a rather homogeneous and low value of $E_{Ra}$, $0.27 \pm 0.04 \text{ Bq kg}^{-1}$, close to the detection limit, with the exception of the “banc royal”, which is less porous and shows a value of $E_{Ra}$ of $2.3 \pm 0.2 \text{ Bq kg}^{-1}$. These beds were quarried as construction materials in the eighteen century, with the “banc royal” as the preferred quality. The centimetric beds located above the Souchet layer show more variable values of porosity, from 12 to 42%, and more variable values of $E_{Ra}$, with an average value of $1.44 \pm 0.19 \text{ Bq kg}^{-1}$ and a SCSD of $48 \pm 10\%$. The roof bed is characterized by a low porosity and a low value of $E_{Ra}$ $(0.15 \pm 0.04 \text{ Bq kg}^{-1})$, similar to the thick beds, which suggests some relation between $E_{Ra}$ and the mechanical strength. By contrast, clay layers, with an average of $4.1 \pm 0.4 \text{ Bq kg}^{-1}$, show significantly higher values than the limestone beds, probably due to the preferential adsorption of radium on clay minerals. This study indicates that the main radon sources in the Vincennes quarry are the clay layers, the filling waste materials stored in the quarry and the “bancs francs” beds.

**Fig. 8.** $E_{Ra}$ of calc-schist samples (left) and of calc-schist and mica-schist samples (right), from the Syabru-Bensi (Central Nepal), versus position along the outcrop. Location (a, c) and $E_{Ra}$ results (b, d) of both outcrops are presented.
3.4. Large-scale geological dispersion for rocks and formation consistency

When the behaviour of $EC_{Ra}$ at scarp level is reasonably well understood, and that one has gained some confidence that values of $EC_{Ra}$ at scarp level are representative of broader geological structures, one may study variations at larger spatial scales, from formation to formation, and address the large-scale dispersion (LASD) of the $EC_{Ra}$ values. One example is given in Fig. 11, showing a profile of $EC_{Ra}$ across the Basal Penninic Contact (BPC), one of the major faults of Alpine tectonics (Ceriani et al., 2001), at the “Roselend Cormet”, located at an altitude of 1968 m near the Roselend lake (Richon et al., 2005; Trique et al., 2002).

![Diagram](image-url)
The $EC_{Ra}$ values, shown from West to East in Fig. 11, overall were rather low, with an average value of $1.4 \pm 0.2$ Bq kg$^{-1}$ and a LASD of 65 $\pm$ 13%, by contrast with other metamorphic rocks, for example from Nepal (see below), where $EC_{Ra}$ in gneisses could reach values larger than 50 Bq kg$^{-1}$. Values here appeared rather homogenous, with smooth variations, an average value of $1.05 \pm 0.14$ Bq kg$^{-1}$ and a LASD of 32 $\pm$ 10%, in the Valaisan domain, east of the BPC. West of the BPC, values appeared more dispersed, with an average value of $1.6 \pm 0.3$ Bq kg$^{-1}$ and a LASD of 67 $\pm$ 16%, with minimum values of 0.19 $\pm$ 0.05 Bq kg$^{-1}$ in the Callovo-Oxfordian and Kimmeridgian domains. Larger and more dispersed values correspond to formations containing organic carbons, such as the Houiller black sandstone, and the shear zones at the level of the BPC and also within the highly deformed and thrusted Permian and Bajocian domains. While the $EC_{Ra}$ value itself is interesting, the LASD seems to be a powerful index to characterize the geological structural heterogeneity. Maxima seem to coincide with recognized faults, bringing additional support to previous evidence of enhanced radon exhalation in the vicinity of faults (Ciotoli et al., 2007; Richon et al., 2010).

4. General properties of effective radium concentration

4.1. Effective radium concentration in soils

Values of $EC_{Ra}$ obtained so far for soils, mostly from Nepal, are shown in Fig. 12 and compared with the values measured for rocks. These distributions, while obtained with our particular samples from particular locations, exhibit some general properties of interest. Values for soils vary from about 1 Bq kg$^{-1}$ for sandy soils to about 50 Bq kg$^{-1}$ for dark and red soils (Girault et al., 2011a, 2011b). Values lower than 1 Bq kg$^{-1}$ are encountered only in exceptional circumstances, essentially soils consisting of almost pure quartz sands. The maximum value so far, 8840 $\pm$ 710 Bq kg$^{-1}$, was observed in red ferric oxides deposits near a spring in the Higher Himalayas of Nepal. Other relatively rare and large values, between 50 and 1000 Bq kg$^{-1}$, originated from granitic and uranium-rich soils in France and from geothermal zones in Nepal. The distribution shows two peaks, representative of the two most frequent situations: brown soils, with a typical $EC_{Ra}$ of 10 Bq kg$^{-1}$, for example representative of most soils in temperate climates, and dark soils with a typical $EC_{Ra}$ of 20 Bq kg$^{-1}$. Considering the whole soil data set, we obtained a median $EC_{Ra}$ value of 11.74 $\pm$ 0.01 Bq kg$^{-1}$. To be able to interpret radon exhalation fluxes from the ground surface, such knowledge of the radon source term is necessary.

4.2. Effective radium concentration in rocks

The overall distribution of $EC_{Ra}$ for rock samples, shown in Fig. 12, spans 6 orders of magnitude, with most commonly found values between 0.1 and 10 Bq kg$^{-1}$, with median $EC_{Ra}$ value of 1.52 $\pm$ 0.01 Bq kg$^{-1}$. In general, the $EC_{Ra}$ of rocks, thus, is smaller than the $EC_{Ra}$ of soils. This fact is due to both a larger mean radium content in soils, owing to reprecipitation during dissolution of the soluble phases, both during silicate and carbonate alteration, and a larger emanation coefficient, probably related to microcracking in the soil minerals and reprecipitated radium more often distributed.
on grains surface than in bulk minerals. The $EC_{Ra}$ of rocks nevertheless can show exceptionally large values, with a conspicuous peak above 100 Bq kg$^{-1}$, for samples collected in the Limousin granite (central France), rich in uranium ore. More interestingly, such large values were also found in gneiss samples from the Lesser Himalayan Sequences in Nepal.

Some general features of $EC_{Ra}$ emerge when studying the distributions of $EC_{Ra}$ values separately for various rock types (Fig. 13). Statistical parameters are given in Table 3 for each rock type, and also given separately for a selection of particular locations.

Larger values are observed for granites, with a 90% probability range from 1.5 to 620 Bq kg$^{-1}$. While conspicuous spikes are visible for $EC_{Ra} > 10$ Bq kg$^{-1}$, some granites, indeed, did not necessarily have large $EC_{Ra}$, in contradiction with the preconception that granitic zones are always radon-prone areas. One may raise the question, however, whether the rocks labelled as granites in geological maps, are systematically genuine granites. For example, the so-called “Royat granite” near Clermont-Ferrand, characterized by $EC_{Ra} = 2.82 \pm 0.37$ Bq kg$^{-1}$, is actually a migmatite associated with magma release, instead of a pluton such as in Limousin. By contrast, volcanic rocks in our data set were characterized by comparatively low values of $EC_{Ra}$ with 90% of samples having $EC_{Ra}$ between 0.09 and 4.2 Bq kg$^{-1}$, with little differences between the considered volcanoes (Table 3). This fact may not be general. Indeed, in Italy, some high radium contents have been reported associated with magma activity (Voltaggio et al., 2004).

Sedimentary and volcanic rocks have similar values of $EC_{Ra}$ in our data set (Fig. 13 and Table 3). Lower values were however reported for the Vincennes Lutetian samples. Sedimentary rocks can have values significantly larger, for example for the Siwaliks samples from Nepal, detritic rocks from highly metamorphosed crystalline and carbonate rocks. This fact might indicate that a long history of fabric, deposition, metamorphism, alteration and redeposition, might progressively lead to enhanced values of $EC_{Ra}$, which, in some way, could indicate the weathering-alteration age of the rock (Schmidt and Cochran, 2010).

The $EC_{Ra}$ values from metamorphic rocks span a wide range from 0.10 to 19 Bq kg$^{-1}$. However, rocks labelled as quartzites do not necessarily have small values of $EC_{Ra}$, with values larger than 5 Bq kg$^{-1}$ commonly observed. These values were thus larger, for example, than the whole set of Roselend gneiss samples.

4.3. Effective radium concentration in building materials

In our sample of construction materials, while the statistics remained small, we note a large variability of $EC_{Ra}$ values (Table 3). While the $EC_{Ra}$ of bricks and cements were consistently small in our
5. Discussion and conclusion

The results presented in this paper support the use of $E_{C_Ra}$ as a powerful and subtle geological parameter and the idea that large-scale measurement efforts might lead to fundamental breakthroughs in understanding of radon emanation and of radium distribution in rocks. Thus, while separating into radium concentration $C_{Ra}$ and emanation coefficient $E$ can be useful to refine our understanding, the product $E_{C_Ra}$ is, by itself, an interesting quantity, rather than just an intermediate number. $E_{C_Ra}$ values indeed showed remarkable coherence at scarp level, with smooth spatial variations, and were overall remarkably representative of the geological formations. Dispersions at sample-scale and at scarp-scale, while moderate, nevertheless could be significant and appeared themselves as important parameters reflecting the nature of the radium distribution. At larger scales, $E_{C_Ra}$ values could show remarkable stability over some geological formations, and heterogeneities and rapid variations, for example in the vicinity of shear zones. Thus, a detailed characterization of $E_{C_Ra}$ might open new perspectives on the physics of uranium mineralization, in particular in relation with active tectonics. However, in general, the coherence of $E_{C_Ra}$ values and the sample-scale, scarp-scale and formation-scale dispersions must be studied carefully in every practical situation.

When looking at the scarp $E_{C_Ra}$ average, this study shows necessary caution when collecting only one sample, which can sometimes only reflect a local anomaly such as remineralisation, reprecipitation or redeposition. It is particularly true for heterogeneous outcrops, as well as outcrops which might seem homogeneous at first glance. Consequently, various samples need to be collected in order to obtain a sufficiently significant $E_{C_Ra}$ average value and its range of variation. Such $E_{C_Ra}$ range is of practical interest in radon exhalation rate and concentration studies and in modelling of radon flux, in both diffusive and advective contexts. With sufficient amount of samples, it might be possible to investigate the statistical nature of the distribution of values. For example, the log-normal hypothesis, which seems to apply to numerous geochemical parameters, could be tested in the case of $E_{C_Ra}$ (Bossew, 2010).

When dealing with sufficiently large numbers of samples, $E_{C_Ra}$ emerges as a promising tool to characterize geological structures from centimetric to kilometrical scales, which may ultimately lead to a better understanding of radium distribution and radium transport, which can also be used as a proxy of potential sensitivity and resilience of natural sites to pollutants. Such endeavours, thus, might be of great interest for environmental applications, much beyond radon hazard prevention, mitigation and control. Furthermore, such detailed investigations give some confidence that radon spatial variations can be interpreted and give hope that, with more efforts, maybe, important temporal variations, such as periodic components (Barbosa et al., 2010; Richon et al., 2009; Crockett et al., 2010) or transient variations potentially associated with earthquake precursory phenomena (Steinitz et al., 2006; Kumar et al., 2009; Ghosh et al., 2009) can be identified in a reliable manner.

Acknowledgements

The authors thank Gauthier Hulot and Edouard Kaminski for continuous and enthusiastic support during difficult times. Jérôme Wassermann is thanked for the PERM4 drill core samples and for discussions on the EDZ of the Roselend tunnel. The quality of the original paper was improved thanks to the careful work of three anonymous reviewers. This paper is IPGP contribution number 3304.


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