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Measuring effective radium concentration with less than 5 g of rock or soil

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ABSTRACT

Radon generation in natural systems and building materials is controlled by the effective radium concentration EC_{Ra}, product of the radium concentration C_{Ra} and the emanation factor E. An experimental method is proposed to measure EC_{Ra} in the laboratory by radon accumulation experiments using less than 5 g of sample inserted in 125 mL scintillation flasks. Accumulation curves with fine temporal resolution can be obtained, allowing the simultaneous determination of the effective leakage rate. The detection limit, defined as the EC_{Ra} value giving a probability larger than 90% for a determination with a one-sigma uncertainty better than 50%, is moderate, varying from 2 to 5 Bq kg^{-1} depending on the conditions. Obtained punctual uncertainties on EC_{Ra} vary from about 10 to 20% at 10 Bq kg^{-1} to less than 3% for EC_{Ra} larger than 500 Bq kg^{-1}. The representativity of small samples to estimate meaningful values at site or system level is, however, a definite limitation of the method, and the sample dispersion needs to be considered carefully in every case. Nevertheless, the value obtained with 5 g or less differs on average by 9 ± 13% from the value given by standard methods using 100 g or more, thus is sufficiently reliable for most applications. When EC_{Ra} is sufficiently large, the temperature sensitivity of EC_{Ra} can be measured reliably with this method, with obtained mean values ranging from 0.39 ± 0.05% °C^{-1} for Compreignac granite, to 2.8 ± 0.2% °C^{-1} for La Crouzille pitchblende, both from the centre of France. This method is useful to study dedicated problems, such as the small scale variability of EC_{Ra}, and in circumstances when only a small amount of sample is available, for example from remote areas or from precious materials such as historical building stones.

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1. Introduction

Radon-222, rare gas and decay product of radium-226 in the uranium-238 decay chain, is released from rocks and soils, and accumulates in the atmosphere of buildings or in poorly ventilated underground settings (Tanner, 1964). An alpha emitter with a half-life of about 3.8 days, radon-222, mainly because of its radioactive daughters, is responsible for half of the annual dose of the general population (Porstendörfer, 1994) and is consequently a major radiation hazard (Al-Zoughool and Krewski, 2009). Radon-222 can also be used as a tracer of geological fluids and, thus, is useful to assess near surface processes (Perrier et al., 2009a; Pascale Tommasone et al., 2011) or crustal processes in volcanic (Giammanco et al., 2007; Eff-Darwich et al., 2008; Laiolo et al., 2012) and seismically active zones (Steinitz et al., 2006; Richon et al., 2011). To assess the radon temporal and spatial distribution (Nazaroff et al., 1985; Perrier and Richon, 2010), however, a prerequisite is a sufficient knowledge of the radon source.

Not all radium atoms present in a mineral lattice are able to release a radon atom into the pore space, allowing its subsequent transport to the considered target, but a fraction E called the emanation factor (Tanner, 1964; Sakoda et al., 2011). The radon source term, thus, is the product of E by the radium concentration C_{Ra}, and is referred to as the effective radium concentration EC_{Ra}, expressed in Bq kg^{-1} (Stoulos et al., 2004). The emanation factor E results from the spatial distribution of radium in mineral grains, the properties of the porous network, and other physical parameters such as temperature (Girault and Perrier, 2011) and moisture content (Sakoda et al., 2010). While some aspects of the related physics are well understood (Barillon et al., 2005; Adler and Perrier, 2009), it remains difficult to predict the value of E, which can vary from less than 1% to values larger than 25% (Markkanen and Arvela, 1992; Bossew, 2003; Sakoda et al., 2011). In every application, consequently, it is necessary to measure EC_{Ra} experimentally.

To measure EC_{Ra} in the laboratory, one method is to perform accumulation experiments by inserting a sample in a closed container and measuring the radon concentration with integrating solid state nuclear track detectors (SSNTD) or, as a function of time, with scintillation cells or automatic radon detectors (Stoulos et al., 2003; Tuccimei et al., 2006; López-Coto et al., 2009). Experiments

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with SSNTD have accumulation volumes of the order of 1 L (Baixeras et al., 2001; Singh et al., 2002, 2007; Ramola and Choubey, 2003; Prasad et al., 2008), but, most of the time, the volume of the accumulation chamber is larger than a few litres: for example about 4 L (Stranden et al., 1984; Heiligmann et al., 1997; Righi and Bruzzi, 2006; Tuccimei et al., 2006), 5 L (Kovler et al., 2005), 7 L (Iskandar et al., 2004), 9 L (Kovler et al., 2005; Hassan et al., 2011; Tetsuo Ishikawa, private communication), 10 L (Chau et al., 2005), 15 L (Chao et al., 1997), 19 to 20 L (Menetrez et al., 1996; Amaral et al., 2012), 26 L (Sakoda et al., 2008), 50 L (Al-Jarallah, 2001; Al-Jarallah et al., 2005), 55 L (Lysandrou et al., 2007), 60 L (López-Coto et al., 2009), 110 L (Stoulos et al., 2003), 220 L (López-Coto et al., 2009), and even volumes of 1 m^3 (Jang et al., 2005) or larger than 2 m^3 (Petropoulos et al., 2002). Such accumulation volumes require, to achieve a meaningful sensitivity, a minimum sample mass at least of the order of 100 g, and more often of the order of 500 g. In some circumstances, however, for example when a large number of samples needs to be transported from remote areas, or in the case of precious samples from space or historical buildings, the available sample mass is smaller than 100 g. To reduce the sample mass necessary for EC_{Ra} measurement, one idea is to use the radon detector itself as accumulation chamber. For example, EC_{Ra} has been measured with 65 g of a Martian analogue soil in a vessel permanently connected to a scintillating flask during the accumulation experiment (Meslin et al., 2011). Accumulation experiments with high temporal resolution (1 h) have also been performed (Ferry et al., 2002) using argillite samples from the Tournemire tunnel (France), with a mass of about 15–30 g, directly attached to an Alphaguard radon monitor (Genitron™, France), able to monitor with sufficient precision radon concentrations smaller than 100 Bq m^{-3}. The accumulation volume in this set-up, however, cannot be reduced below 960 mL, volume of the detection chamber in the Alphaguard, preventing measurements with smaller masses. Furthermore, laboratory experiments with costly sensors such as the Alphaguard cannot be performed routinely and, consequently, less than 10 measurements have been reported so far with this method (Ferry et al., 2002; Richon et al., 2005).

In this paper, we propose a method to measure EC_{Ra} with a mass smaller than 5 g of soil or rock sample, by directly inserting the sample in a 125 mL scintillation flask, which is then used as the accumulation chamber. Examples of obtained accumulation curves are shown and interpreted to illustrate the potentials and limitations of this method. The measurement uncertainties and the sensitivity levels are discussed using experimental data and Monte-Carlo simulations. Then, we analyse in which respect the results, obtained using this method, are meaningful, and we give examples of applications.

2. Method and sample installation

Scintillation flasks (Lucas, 1957) from Algade (France) with 125 mL volume are used in this study (Fig. 1). The neck of the flask, not covered with the ZnS(Ag) coating, having a diameter of 25 mm and a length of 30 mm, can be used as a storage volume (14.7 cc) for the sample to be measured. To avoid contributions from thoron (radon-220, from disintegration of radium-224), the sample, with mass m, measured with a SARTORIUS™ BP11D balance regularly checked to a precision of ±0.005 g, is wrapped in three layers of folded filter paper and inserted in a small cotton bag, attached to the rubber stopper of the flask by two needles (Fig. 1). After the flask is closed, the air tightness is checked by depressurization and atmospheric pressure is restored by inserting a syringe needle. Accumulation time zero is defined as the moment the syringe is removed.

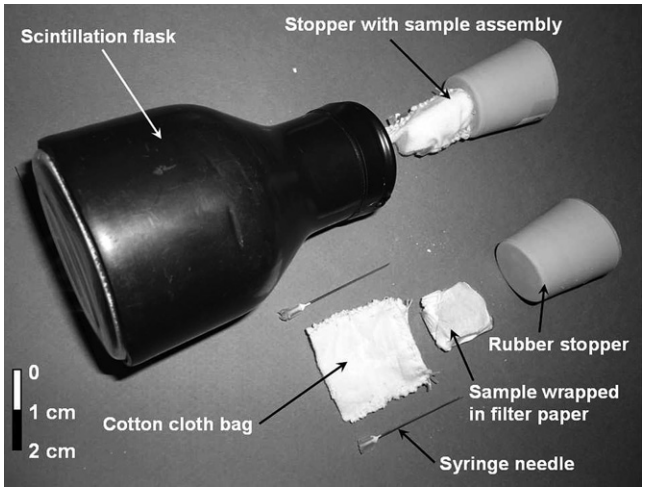

Fig. 1. Picture of the installation scheme of the sample in the 125 mL scintillating flask.

Radon accumulation is subsequently measured by inserting the flask into a CALEN™ (Algade, France) photomultiplier. The radon activity concentration in Bq m^{-3}, at a certain time, is obtained by averaging two counts, with counting time varying, in our experiments, from one to 10 min, and multiplying the counting rate per minute by an absolute calibration factor of 71.3 ± 3.6. Five CALEN™ photomultipliers are used in this study, among which the dispersion of the calibration factor, checked regularly, is always smaller than 2%.

Given the small amount of sample material, the determination of the smaller values of EC_{Ra} depends dramatically on the knowledge of the properties of the scintillation flasks, and primarily, their intrinsic background counts in the absence of sample. The background counts of 76 flasks have been monitored over several years (see Appendix A). Background counts appear to be an intrinsic property of each flask, relatively stable with time, irrespective of the history of its use in the laboratory or in the field. Some flasks have small background counts, as low as 0.2 count per minute on average, and some have larger background counts, as large as 1–2 counts per minute on average, thus inappropriate to make proper accumulation experiments. Background counts of the flasks are in any case measured before the installation of the sample and are subtracted from the subsequent counts during accumulation to obtain the radon concentration. This background subtraction also takes care of potential residual radon concentration in the atmosphere of the laboratory.

Scintillation flasks closed by pre-perforated natural rubber stoppers are not totally air-tight. Their leakage rate can be measured in dedicated experiments, for example, in the field, at locations with gas discharges providing large radon concentrations (Perrier et al., 2009b). The decay rate of the counting rate of each flask can be followed as a function of time, and shows an exponential decay rate with a decay constant λ_{exp} larger than the radon-222 decay constant λ (2.1×10^{-6} s^{-1}). The normalized leakage (or ventilation) rate a_V, defined as $a_V = \lambda_{exp}/\lambda - 1$, is found to vary from 0.02 to 0.33, with an average value of 0.08 ± 0.02. Another experiment provided values varying from 0.02 to 0.24, with an average value of 0.13 ± 0.09.

The absolute calibration constant can also vary from flask to flask. This effect has been studied in the field at a location where radon discharge was found to be sufficiently constant with time using autonomous continuous sensors (Richon et al., 2011). At one location, 37 flasks were sampled and their counting rates were compared to provide an intercalibration factor for each flask. The experiment, repeated one year later, provided consistent values of

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