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New study on the correlation between carbon dioxide concentration in the environment and radon monitor devices



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ABSTRACT

The influence of high geogenic carbon dioxide concentrations on monitoring devices might present a significant challenge to the measurement of radon concentrations in environments with a high level of carbon dioxide concentration such as volcano sites, mofettes, caves, etc. In this study, the influence of carbon dioxide concentration on several different types of radon monitor devices - including Alpha Spectrometry (Sarad RTM 2200, EQF 3220, RAD7), Ionizing Chamber (AlphaGUARD PQ2000 PRO) and Active Cell (Active scintillation cell, Pylon 300A) — was examined to represent new aspects of radon measuring in environments with carbon dioxide. In light of the results, all measuring devices were exposed to variable conditions affected by carbon dioxide concentration, except for the AlphaGUARD, which was kept in a steady state throughout the experiment. It was observed that alpha spectroscopy devices were affected by carbon dioxide, since measured radon concentrations decreased in the presence of 70% and 90% carbon dioxide concentrations by 26.5 \pm 2% and 14.5 \pm 2.5% for EQF 3220, and 32 \pm 2% and $35.5 \pm 2\%$ for RTM 2200. However, the ionizing chamber instrument was unaffected by changes in carbon dioxide concentration. It was determined that the RAD7 performed relatively inefficiently in the presence of carbon dioxide concentrations higher than 67% by an overall efficiency factor of approximately 0.52, confirming that it is not an admissible radon monitor instrument in environments with high carbon dioxide concentrations.

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1. Introduction

Radon (Rn-222) emanation measurements are frequently performed in geological and environmental studies where high concentrations of carbon dioxide are often found in soil, caves, mofettes (natural CO₂ spring areas) or fumaroles adjacent to volcanoes (Bonforte et al., 2013; Zimmer and Erzinger, 2003; Phuong et al., 2012; Heiligmann et al., 1997; Pérez et al., 2007; Papp et al., 2010). In this context, it should be mentioned that in the presence of carrier gases such as methane (CH₄) or carbon dioxide (CO₂), the radon flux might be larger than in other environments (Martini et al., 2010; Frunzeti et al., 2013). The theory of this phenomenon states that deep, magmatic sources are characterized by high radon activities and high CO₂ effluxes, while shallow sources are indicated by high thoron activities and rather low CO₂ effluxes (Giammanco et al., 2007). However, the importance of this theory arises from the fact

* Corresponding author. E-mail address: ashahrokhi@almos.uni-pannon.hu (A. Shahrokhi). that certain mofette sites (natural CO₂ springs), characterized by an elevated level of CO₂, can be used as monitoring, assessment and forecasting tools in geographical studies, e.g., in monitoring faults; to assess the potential risks of a volcano, using CO₂ as a tracer; to forecast a volcano's potential, etc. (Moloney et al., 2011; Saurer et al., 2003). Escaping CO_2 from deep underground contains an amount of natural radioactive Rn-222 gases that it is important to monitor, not only due to radiation dosimetry aspects, i.e., a strong correlation between radon exposure and lung cancer occurrence (Somlai et al., 2011), but also for geographical studies, i.e., for a better understanding of radon exhalation associated with tectonic or volcanic processes (Richon et al., 2005). Consequently, it should be mandatory to monitor the radon levels of mofettes used for curative purposes due to high level of the radon activity concentration in the mofettes' indoor air and dosimetery is needed to estimate the effective dose received by workers and patients (Néda et al., 2008a, 2008b). Lane-Smith and Sims (2013) have recently shown that the active Rn-220 and Rn-222 measuring devices could be affected by the presence of CO₂ while performing radon or thoron measurements. They noted that for every percentage of CO₂ concentration, the calculated Rn-220 concentration should be multiplied by a correction factor of 1.019, whereas the Rn-222 concentration ought to be multiplied by 1.003, on account of Lane-Smith and Sims' observations regarding the effects that CO_2 might exert on radon measurements performed with radon monitor devices used worldwide (Lane–Smith and Sims, 2013).

In the present study, a new experiment was carried out to determine the influence of variable carbon dioxide concentrations on the most common types of radon monitor devices available on the market.

2. Materials and methods

In order to analyse the potential effects of environmental carbon dioxide on the radon monitor devices, a system was assembled to facilitate the use of a controllable Rn-222 and CO₂ concentration in a confined space. A newly developed and certified 210.5 L metal radon chamber (Genitron EV 03209) with a removable lid, and an electric fan in order to ensure internal homogenization, was used as an experimental chamber. The carbon dioxide was fed to the chamber through a system of tubing connected to a flow meter and a CO₂ sensor, which continuously monitored the CO₂ level inside the chamber. The gas intake flux was kept constant, at one litre per minute. A certified radon source (Pylon RN2000A, a passive radon gas source) supplied a known concentration of radon through another system of tubing within the chamber in which the experiment was conducted. The passive radon source (Pylon Electronics Inc., Canada) used for this purpose was 105.7 + 0.42 kBg radium (²²⁶Ra) covered in a leak-proof metal vessel to emanate radon gas at a constant rate. After the criteria for a muffed atmosphere were met, four different devices commonly used for monitoring radon were introduced inside the above-mentioned chamber. The four devices used in the test were chosen because they involve different measuring methods, namely: alpha detector devices (RTM 2200 and EQF 3220, Sarad, Germany), RAD7 (Durridge Company, Inc.) and an ionizing chamber device (AlphaGUARD PQ2000 PRO, Saphymo Gmbh, Germany).

AlphaGUARD was set up in diffusion mode to conduct measurements over a period of 10 min, while RAD7 was set up with the following settings: protocol User, Mode of sniff, 10 min of cycling, unlimited recycles, Thoron off, Pump Auto and tone off. A drying tube (CaCl₂) was installed on the inlet of RAD7 to keep the humidity inside the RAD7 container below 10%, in line with the user manual's guide. Both EQF 3220 and RTM 2200 were set to sample for 10 min on fast mode while these devices measured the CO₂ concentration in the air. Then all the devices were placed in the chamber, which was sealed and vacuumed. Then the Pylon 2000A passive radon gas source was connected to the chamber to reach a specific activity of approximately 20 kBq/m³, whereas the radon activity concentration inside the chamber was kept constant for the entire duration of the experiment by use of a three-way connector Y joint with a regulator and a flow meter connected to the outlet of the radon source chamber and the inlet of the experiment chamber, respectively. Meanwhile, the connector's third connection was attached to the outside by a plastic pipe. Then, by controlling the flow rate using a flow meter and a regulator, the radon concentration inside the experiment chamber was kept constant with an uncertainty value of 0.2 kBq/m^3 . Fig. 1 presents a schema of the experimental system.

Lastly, the carbon dioxide was injected into the chamber to increase its concentration from 0% to approximately 89%. After each sampling and measurement, the carbon dioxide concentration was increased while the radon concentration was kept at the level stated 20 kBq/m³. The experiment was separated into the two phases; in the first part, the carbon dioxide concentration was increased from 0% up to 17%; however, in the second phase, the CO₂



Fig. 1. Schematic of experiment system.

concentration inside the chamber reached 67%, and then increased by 5% after each sampling period to 89%. In both phases, the concentration of the reference radon was about 20 kBq/m³. Simultaneously with each significant increase in the CO₂ concentration, different radon samples were grabbed from both inside and outside the chamber. A fifth device based on grab sampling into a 272 ml Active Cell (Pylon 300A Lucas type scintillation cell, Pylon Electronics Inc, Canada) was used to take gas samples from the experimental chamber. According to the number of counts provided by Pylon, the radon activity concentration was later calculated using the following Equation (1):

$$\mathbf{R}_{\mathbf{c}} = (\mathbf{C} - \mathbf{B}) / (\mathbf{E} \, \mathbf{S} \, \mathbf{3} \, \boldsymbol{\varepsilon} \, \mathbf{t} \, \mathbf{V}) \tag{1}$$

where R_c is radon concentration (Bq/m³), C is number of counts, B is the background count, E is efficiency of the scintillation cell, ϵ is the efficiency of sampling (0.98), V is the volume of the cell (0.27 \times 10⁻³ m³), t is the measuring time (Second), 3 is the number of alpha emitting radionuclides after four hours equilibrium (radon and its two short-lived decay products), and S is a correction factor which is calculated using S = e^{-\lambda t} where λ represents the decay constant.

To maintain the activity concentration of radon inside the chamber, the total daily radon emanation rate from the radium source was calculated using Equations (2)-(4); then, by adjusting the flow rate, the radon concentration remained constant during the experiment.

$$R_{Em} = A_0 e^{-\lambda t} \tag{2}$$

$$\lambda = ln(2)/T \tag{3}$$

$$R_{c} = A/V \tag{4}$$

where R_{Em} is the daily radon emanation from the source (Bq), A_0 is the initial activity that was provided by the producer (Bq), λ is the decay constant which is calculated from Equation (3), t is the time

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