COMBINED LSC-BASED METHOD FOR RADON IN AIR MEASUREMENTS

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ABSTRACT. A combined method for ^{222}Rn measurement in air based on liquid scintillation counting (LSC) is proposed. It is an integral and a highly sensitive method, capable for different ^{222}Rn measurement applications. The first method is based on a charcoal trapping. The second method (Kaihola et al. 1992; Kaihola 1996) is based on a plastic scintillation material (Meltilex^{TM})-coated scintillation vial. This method is a scintillation vial method for ^{222}Rn measurements like the Lucas chamber method. Both methods utilize modern LSC techniques and PTFE vials. One essential aim was to perform *in situ* calibration of the charcoal-based method for any weather conditions. We tested the method on both a Quantulus 1220 and a Triathler counter. The corresponding sensitivity is ~0.1 cpm per Bq m^{-3} for 1 day of exposure.

INTRODUCTION

Different methods were used for ^{222}Rn in air measurements: integral continuous measurement methods and short-period measurement methods. Integral methods are based on the application of etch track detectors, electret detectors, or charcoal detectors, while short-period measurement methods use scintillation and semiconductor detectors or an ionization chamber. They are both active and passive types. When selecting an appropriate method one should take into account its peculiarities: sensitivity, minimum exposure time required, dynamic range, impact of humidity and dust, etc. A major goal of this investigation was to perform *in situ* calibration of the charcoal-based ^{222}Rn measurement method for its use under variable weather conditions for air from different sites (mines, open pit, caves, etc). The combined method described here includes the charcoal method and the Lucas chamber method for calibration.

METHODS

The combination of 2 methods for measuring radon in air is proposed here. The first is a charcoal-based method using liquid scintillation counting (LSC) technology, see Figure 1 (Buzinny 1996), and the second adds the use of a scintillation vial (Lucas chamber) method based on LSC (Kaihola et al. 1992; Kaihola 1996; Zelensky and Buzinny 1993). A set of base measurements carried out at one site, at close distance, at the same time, and under the same supposed weather conditions is calibrated using an additional method. The necessity of an additional method is caused by the huge dependence of sensitivity of the charcoal-based method on conditions of charcoal in the surrounding air.

Charcoal Method Based on LSC

A description of the charcoal method based on LSC can be obtained from the work of Buzinny (1996). It includes Rn sorption while sampling in sites, Rn desorption into the LS cocktail, and LSC measurement performed in the laboratory. It allows the repeated use of each element in the entire system. As shown in Figure 2, 3 tablets of charcoal for each cycle of Rn measurement were applied. As tabulated charcoal is ready for use, it is highly convenient for sampling in the field. The low consumption of materials like charcoal and LS cocktail additionally minimizes the costs for analysis. As clearly demonstrated in Figure 3, the PTFE vial gives a better spectra resolution.

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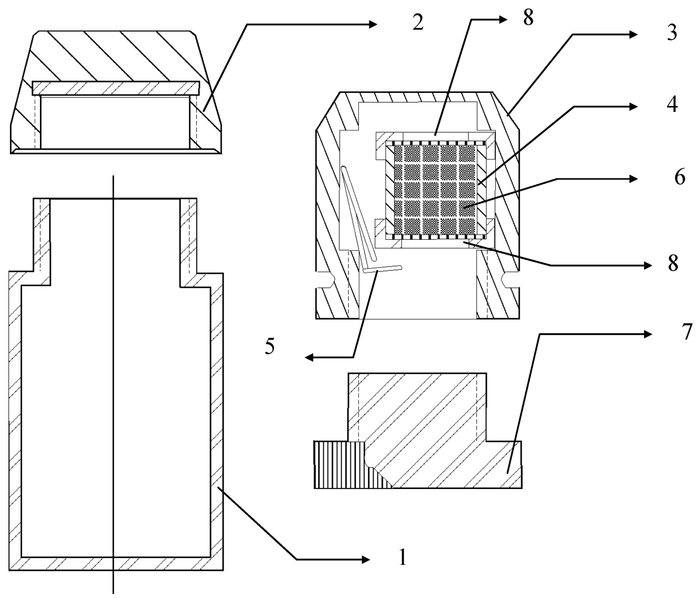

Figure 1 Application of a 20-mL PTFE vial with charcoal canister for measuring radon in air: 1) 20-mL PTFE vial; 2) cap for vial; 3) charcoal canister; 4) charcoal container; 5) metal spring-holder; 6) charcoal; 7) cap of canister; 8) metal mesh (Buzinny 1996).

Figure 2 Set of LS vial, charcoal canister, and charcoal tablets

Scintillation Vial Method

The scintillation vial method based on LSC was developed by Kaihola and colleagues (Kaihola et al. 1992; Kaihola 1996). Direct counting enables high reproducibility and high precision. This method can be carried out quickly and easily. It is based on counting of alpha and beta particles in a thin layer of meltable scintillator, Meltilex^{TM} (Kaihola et al. 1992; Kaihola 1996; Oikary et al. 1991; Suontausta et al. 1993), which covers the walls and bottom of the scintillation vial. Spectral resolution for Meltilex^{TM} is, however, less pronounced in comparison with conventional LS cocktails (see Figure 3 and Figure 4). PTFE vials or polyethylene-coated PTFE vials are more suitable for that specific use. Modern liquid scintillation spectrometers used to measure alpha/

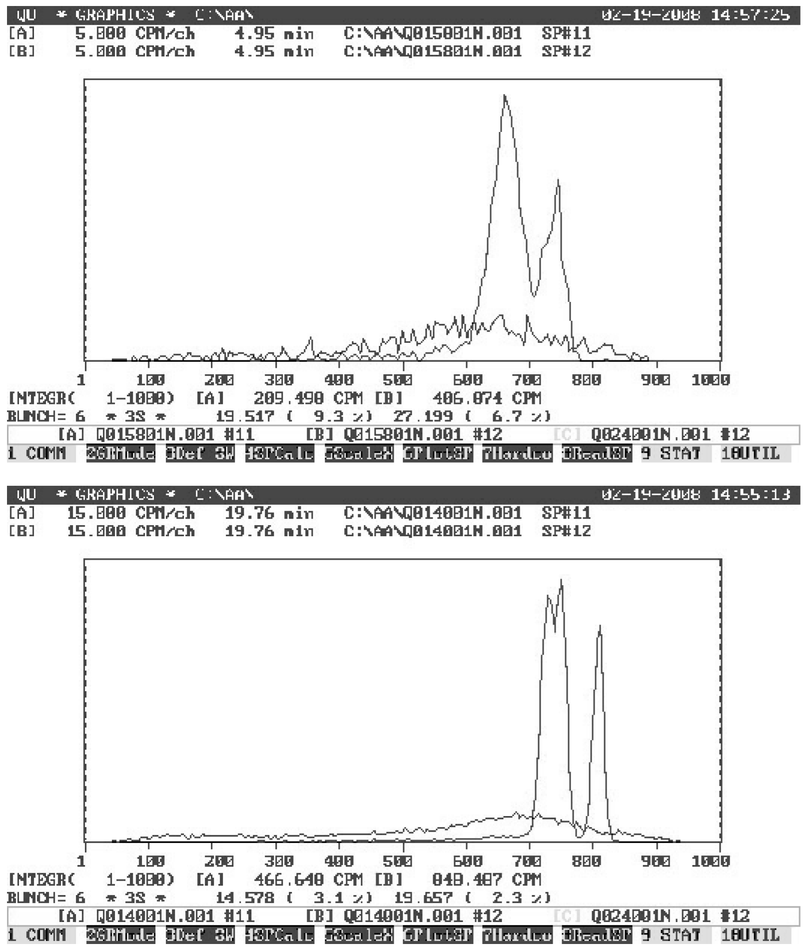

Figure 3 ^{222}Rn in equilibrium with daughters: α and β spectra measured in plastic (upper) and PTFE (lower) vials using a toluene-based LS cocktail.

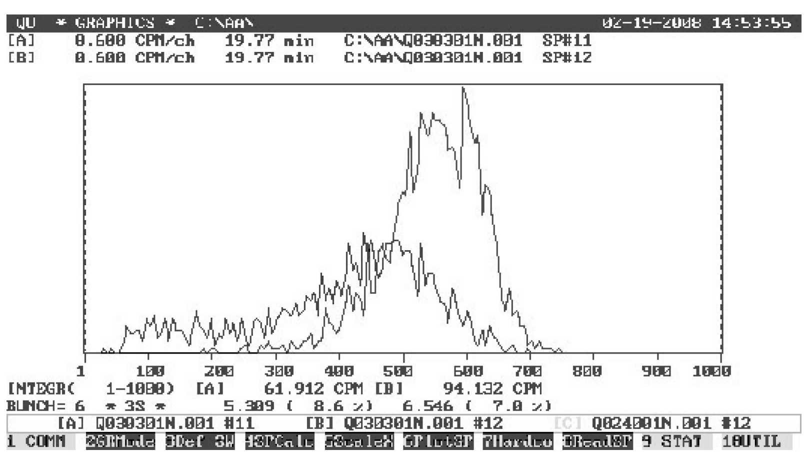

Figure 4 α and β spectra of ^{222}Rn and its daughters measured using a Quantulus 1220 in Lucas chamber (PTFE vial coated with Meltilex™).

beta separation enable Rn measurement via its alpha activity in such vials. The counting efficiency for alpha particles emitted by ^{222}Rn in secular equilibrium with its daughters yields up to 178%. The main disadvantage in the application of this method is the small volume of the LS vial (22 mL). Another drawback is a possible Rn leakage due to imperfect air tightness in the lid.

In situ calibration is carried out by simultaneous measurement of ^{222}Rn in air via the 2 mentioned methods. The measurements for calibration were carried out where the ^{222}Rn in air concentration was relatively high. ^{222}Rn values determined by the relative method are used as a reference for the calculation of relative counting efficiency under various weather conditions. All measurement results, carried out at close measurement points and at similar weather conditions, need correction to correspond with the values of the relative counting efficiency determined.

The activity must be corrected for the total delay due to charcoal canister transportation and vial storage before counting, according to

$$A = A_0 \times \exp(-\ln(2) \times T_d/T_{1/2}) \quad (1)$$

where A_0 is the measured ^{222}Rn in air activity at the site when the sample was collected, T_d is the time between sampling and measurement, and $T_{1/2}$ is ^{222}Rn half-life (3.8235 ± 0.0004 d).

The observed acceptable time for charcoal canister exposure is 1 or 2 d. An advantage of the method described here is its integrating approach. The method has a limit of detection of 1 Bq m^{-3} for 1 d of exposure time and 20 min of counting time, corresponding to a sensitivity of 0.1 cpm per Bq m^{-3}.

The stable counting efficiency of the scintillation vial method allows its application for *in situ* calibration, which can be improved when 2 measurements are performed both at the beginning and at the end of charcoal canister set exposure. The relatively low sensitivity of the auxiliary method is caused by the small volume of the counting vial (~22 cm^3). It is the only limiting factor for site measurements where *in situ* calibration could be simply performed, especially where the ^{222}Rn concentration is above 500–1000 Bq m^{-3}. It should be mentioned that for calibration purposes, all other methods used for routine ^{222}Rn in air measurement require an ^{222}Rn concentration in the range of 2000–10,000 Bq m^{-3}.

Practical Remarks

The PTFE vial has a much better spectral resolution (see Figure 2). The background count rate is also lower when a PTFE vial is used. Medical packaging preserves charcoal tablets for long time in stable active conditions, allowing their application for measurement of radon in air.

A closed charcoal canister keeps ^{222}Rn without leakage during transportation from the site to the laboratory. These days, though, mobile LSC equipment capable of α/β separation enable the possibility to provide *in situ* LS measurements in the field. However, the sensitivity is much better when laboratory LSC equipment is used.

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