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Uncertainty evaluation in radon concentration measurement using charcoal canister

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HIGHLIGHTS

- Measurement uncertainty budget for radon activity concentration established.
- Three different methods for ROI selection are used and compared.
- Recommend to use one continuous ROI, less sensitive to gamma spectrometry system instabilities.

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ABSTRACT

Active charcoal detectors are used for testing the concentration of radon in dwellings. The method of measurement is based on radon adsorption on coal and measurement of gamma radiation of radon daughters. The contributions to the final measurement uncertainty are identified, based on the equation for radon activity concentration calculation. Different methods for setting the region of interest for gamma spectrometry of canisters were discussed and evaluated. The obtained radon activity concentration and uncertainties do not depend on peak area determination method.

1. Introduction

Radon levels in houses and in workplaces have been investigated and monitored worldwide very extensively in the last four decades. The charcoal canister method of radon concentration estimation is the most widely used method of screening (Eremić-Savković et al., 2002; Manic et al., 2006; Tondeur et al., 2011; Yu et al., 1992). This method is sufficiently precise and shows significant correlation with results from etch track detectors (Cortina et al., 2008).

Almost all published data for radon concentration obtained by charcoal canister measurement are without measurement uncertainty. The influence of counting statistics as a major part of the uncertainty was indicated in Zhukovsky et al. (2010) and the total uncertainty of radon concentration measurements conducted by a single charcoal canister was estimated to be between 25% and 35% ($k=2$). Many factors can influence the combined uncertainty of radon measurements. One example is the influence of exposure conditions (Ronca-Battista and Gray, 1988).

In this paper we identify all the contributions to the total measurement uncertainty.

An integral part of the charcoal canister method is the gamma spectrometry of canisters. We show that spectral region of interest (ROI) in gamma spectrometry does not influence the measurement uncertainty.

2. Material and methods

Active charcoal detectors are used for radon concentration screening in dwellings in accordance with the US EPA protocol 520/5-87-005 (Grey and Windham, 1987).

The measurement method is based on radon adsorption on charcoal and measurement of gamma radiation of radon daughters (^{214}Pb and ^{214}Bi). It is based on the high affinity of activated charcoal for several gases and vapors, one of which is ^{222}Rn. Activated charcoal is used to construct passive radon charcoal canisters in the following way: a metal can is filled with low-activity activated charcoal. A metal grid, with 30–50% of the total area in holes, is placed over the charcoal. The can is closed with a padded metal lid, and adhesive vinyl tape is used to seal the canister.

The canister is exposed by removing the metal lid. The exposure should last no less than 48 h. During the exposure, radon is adsorbed to and desorbed from the activated charcoal, and it also undergoes radioactive decay. After the exposure, the canister is re-sealed by replacing the metal lid and vinyl tape. Measurements are then made

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of the radon progeny by gamma spectrometry, with the canisters being placed centrally on the face of the detector. Three gamma energies are used, 295 keV and 352 keV (energies of ^{214}Pb photons) and 609 keV (energy of ^{214}Bi photon). It is necessary to wait at least 3 h between sealing the canister and measurement, in order to attain radioactive equilibrium.

Before every exposure, canisters are prepared by heating open canisters four hours at 110 °C. During the drying, adsorbed water vapor, radon and its progeny, as well as other adsorbed species, are desorbed. After the drying, canisters are sealed and are ready for the next exposure.

3. Calculation of radon concentration

Carbon filters were opened and exposed in closed rooms for between 3 and 6 days and are placed at distance of at least 1 m from the floor and walls. At the end of exposure and after achieving equilibrium between radon and its daughters (at least 3 h after exposure), the measurements were carried out using NaI or HPGe detectors. Radon concentration A_{Rn} is calculated using the equation:

$$A_{Rn} = \frac{G - B}{tE_f C_f D_f} \quad (1)$$

where:

- G gross counts of 3 full energy peaks (295 keV, 352 keV, and 609 keV);
- B gross counts of the same areas of the background spectrum;
- t measurement time (30 min);
- E_f detector efficiency;
- C_f calibration factor for radon adsorption rate;
- D_f decay factor from the midpoint of exposure to the time of counting.

The detector efficiency was determined using a standard canister with the same geometry and matrix as the canisters used for measurements. Standard canister contains a known activity of ^{226}Ra. Background canister is a regular radon measurement canister, which has never been exposed. Detectors were calibrated by ^{226}Ra of known activity in the same geometry (Radium nitrate uniformly distributed in charcoal, Isotope Products Laboratories, Los Angeles, USA). Typical ^{226}Ra spectra obtained with a standard canister are shown on Figs. 1 and 2.

The gross count will depend on the selection of Region of Interest (ROI). In this paper, three different methods for ROI selection are used, and the results obtained by different methods are compared:

1) ROI covers the whole spectral area between 289 keV and 615 keV for HPGe detector (ORTEC) (Fig. 1a) and the whole spectral area between 270 keV and 720 keV for NaI (Fig. 2a). These ROIs include the three peaks of interest.
2) ROI is divided into three parts, where each part represents, respectively, one of the three peaks of interest; peak area is selected by the user (Figs. 1 and 2b).
3) ROI is divided into three parts, where each part, respectively, represents one of the three peaks of interest marked automatically by the software, Canberra–Genie 2 K or ORTEC-Gamma Vision 32 (Fig. 1c).

The Ortec and Canberra spectra and window settings are very similar. For the NaI spectrum the same method was used but, because of the poorer resolution, the peak areas selected by the user were 270–318 keV, 320–415 keV, and 510–720 keV.

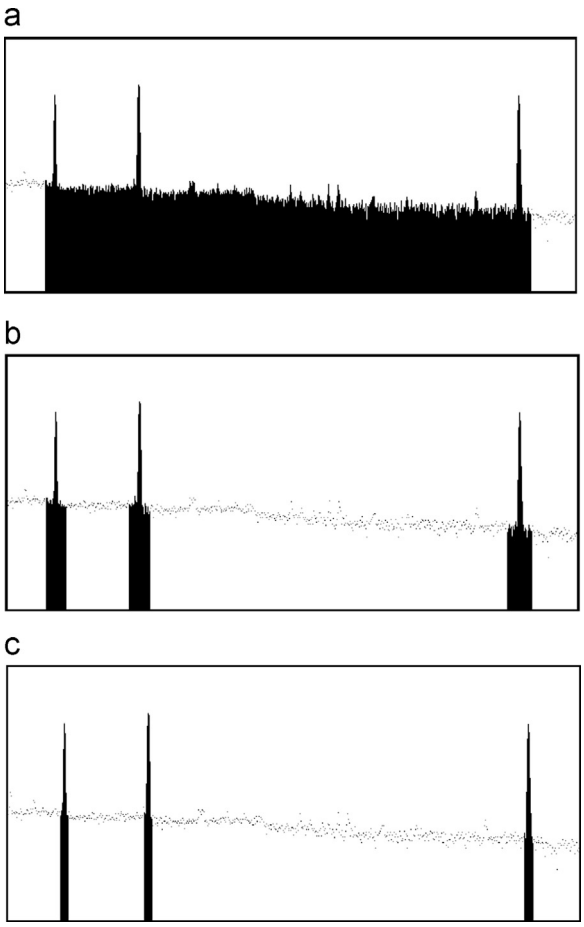

Fig. 1. Spectrum of ^{226}Ra measured with Ortec HPGe detector. Range of interest: (a) the total sum of counts in all channels between 289 keV and 615 keV; (b) peak area selected by the user; and (c) peak area selected automatically by the software (Gamma Vision 32).

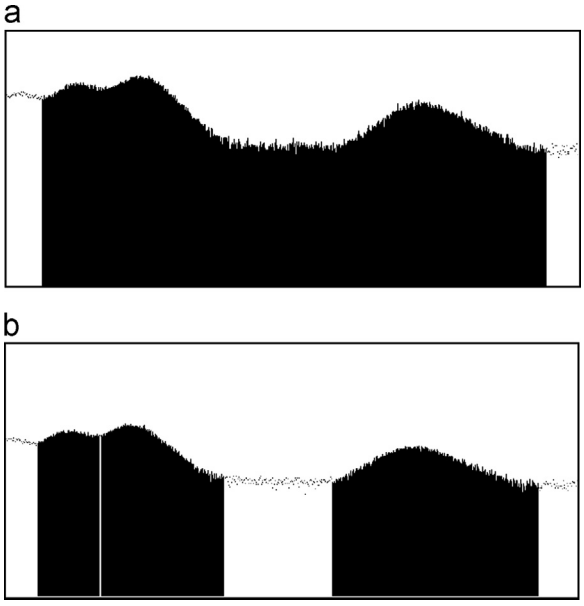

Fig. 2. Spectrum of ^{226}Ra measured with NaI detector. Range of interest: (a) the total sum of counts in all channels between 270 keV and 720 keV; (b) peak area selected by the user (270–318 keV, 320–415 keV, and 510–720 keV).

Methods 2 and 3 require a modification of Eq. (1). While the net peak area can be calculated simply as $G-B$ when there is only one ROI, in case of three separate ROIs, a slight modification is