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Open charcoal chamber method for mass measurements of radon exhalation rate from soil surface

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ABSTRACT

Radon exhalation rate from the soil surface can serve as an important criterion in the evaluation of radon hazard of the land. Recently published international standard ISO 11665-7 (2012) is based on the accumulation of radon gas in a closed container. At the same time since 1998 in Russia, as a part of engineering and environmental studies for the construction, radon flux measurements are made using an open charcoal chamber for a sampling duration of 3–5 h. This method has a well-defined metrological justification and was tested in both favorable and unfavorable conditions. The article describes the characteristics of the method, as well as the means of sampling and measurement of the activity of radon absorbed. The results of the metrological study suggest that regardless of the sampling conditions (weather, the mechanism and rate of radon transport in the soil, soil properties and conditions), uncertainty of method does not exceed 20%, while the combined standard uncertainty of radon exhalation rate measured from the soil surface does not exceed 30%. The results of the daily measurements of radon exhalation rate from the soil surface at the experimental site during one year are reported.

1. Introduction

One of the important indicators of geogenic radon potential of the territory (and, in particular, of construction sites) is the radon exhalation rate (radon flux) from the soil surface. In addition, this information can be used in meteorology for the purpose of monitoring of the global transport of air masses (Arnold et al., 2010) and more accurate weather forecasting, as well as the analysis of the behavior of the global ionosphere (Zhang et al., 2011).

The most common method of measurements of radon exhalation rate from soil surface in the countries of Europe and America is based on the principle of radon accumulation in a closed container (usually cylinder shape, with diameter of 0.1–0.5 m at a volume of 1–10 L), which is mounted on the surface of the soil (IAEA, 2013). Measurement of radon accumulating in the container is carried out in different ways, using (a) electrets (Kotrappa et al., 1993), (b) radon radiometers with either active (Lehmann et al., 2003) or passive (López-Coto et al., 2009) sampling, or (c) activated charcoal, followed by measuring the activity of gamma radiation progeny of radon which accumulated in the activated charcoal (Dueñas et al., 2007).

Recently published international standard (ISO 11665-7, 2012) gives guidelines for estimating the radon-222 surface exhalation rate over a short period (a few hours), at a given place, at the interface of the medium (soil, rock, laid building material, walls, etc.) and the atmosphere. This standard addresses the measurements in the field of on-site investigations, such as the search of radon sources or comparative studies of exhalation rates on the same site. This method is based on the accumulation of radon gas in a closed container.

During the summer of 2008 the comparative measurements of radon exhalation rate using different radon radiometers and sampling tools, but all based on the same method of radon accumulation in a closed container, were conducted in Spain at four different sites (Grossi et al., 2011). The results of this inter-comparison showed quite satisfactory agreement. However, the conditions of these tests, as the authors pointed out, were specially chosen as most favorable for having stable results – dry soil, positive temperatures, stable weather, and finally the absence of an abnormally high radon flux in these sites. It has to be emphasized that the

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analysis of method uncertainty must be performed over the entire range of possible conditions under which the method may be used in practice; this principle is especially important if the measurements are carried out in field conditions. For this reason, the ISO standard 11665-7 (2012) imposes restrictions on the duration of exposure of the container by 1–3 h. This limitation is introduced due to the fact that the longer is the exposure to radon, and the shorter is the container height, according to Fick's first law, the higher resistance to radon diffusion from the ground is created. This phenomenon is well known; it changes the conditions of free radon diffusion from the soil surface. These two influencing factors, time and geometry, can be even more important when the conditions on site are unfavorable, for example, when the radon migration in soil is dominated by convective transport and high soil humidity.

The ISO standard (ISO 11665-7, 2012) addresses additional factors causing a disturbance in the free surface exhalation rate, which can significantly influence the final estimations:

a) The variations in conditions (pressure, temperature, humidity) inside and outside the accumulation container: To minimize these effects, accumulation is specified in the standard to take place over a period of time with little variation in the external and internal container conditions (heavy rain and showers shall be avoided). However, the accumulation container may be thermally insulated.
b) Inadequate air tightness (leakages) and back diffusion induce radon loss. To minimize the effect of leakages, improving air tightness is recommended. To minimize the effect of back diffusion, the container should be purged with radon-free air before beginning the accumulation process, and the calculation of the exhalation rate should be based on the initial slope of the curve of accumulation.
c) The significant activity concentration of thoron in the soil pores.

Finally, the ISO standard (ISO 11665-7, 2012) provides the algorithms for estimating radon exhalation rate for different methods of measuring radon concentration in the container. However, as far as the implementation of these estimates is concerned, the standard procedure does not guarantee the reliability and accuracy of the measurements, because this standard does not define the calibration procedure of the measurements of radon surface exhalation rate.

To the best of our knowledge, the issues of metrological assurance of the measurements of radon exhalation rate from the soil surface under natural field conditions have not yet been discussed in the literature. It should be noted that some authors did conduct similar studies (Kotrappa et al., 2004; Alharbi and Akber, 2014), but the methodology and the results obtained in these studies are not universal and do not allow calibration of a variety of methods and tools for measuring radon surface exhalation rate, and assessment of the results uncertainty considering unfavorable measurement conditions. For example, in the study by Kotrappa et al. (2004) the accuracy of radon surface exhalation rate measured only from the solid surface of building materials in the laboratory conditions is discussed; while the work by Alharbi and Akber (2014) focuses mainly on the effectiveness of the sorption layer of charcoal in the closed accumulation container depending on the thickness of this layer and also on the duration of sampling – again under favorable laboratory conditions only. Therefore, it seems to be highly relevant to discuss not only the principles, but also the details of metrological support of radon exhalation rate measurements from the soil surface, which must be universal for all different known measurement methods and sampling tools, as well as to provide the ability to test this characteristic under in both favorable and unfavorable measurement conditions (sampling).

At the same time, in the mid-1990s, a method for measuring radon exhalation rate from the soil surface using an open chamber with activated charcoal was developed in Russia (Zabolotsky, 2005). Thanks to the open design of the chamber and use of activated charcoal, this method causes very little disturbance in the free radon exhalation rate of the soil surface, in comparison with the closed container method. In order to uncertainty assessment of radon surface exhalation rate (or calibration), special metrological equipment was developed and the open charcoal chamber method was tested in different conditions of sampling, including under unfavorable conditions. The method of measurement of radon exhalation rate from the soil surface using the open charcoal chamber became the most popular method in Russia since 1998; it is widely used nowadays as a part of engineering and environmental studies to evaluate the potential radon risk of building sites in this country.

2. Methods

2.1. Principle and description of the open charcoal chamber method

The principle of the open charcoal chamber is in the absorption of radon atoms exhaling from the limited soil surface by the activated charcoal layer, located in the accumulation open chamber, the scheme of which is shown in Fig. 1.

Before mounting the accumulation chamber on the soil to be tested (Fig. 1), the charcoal is removed out of the working sorption column (Fig. 2) into the accumulation chamber and forms the working layer; while the protective casing sorption column filled with the same charcoal is inserted into the neck of the chamber forming a protective layer (two sorption columns are used to prepare one accumulation chamber). The protective layer prevents radon absorption by the working layer from the near-surface air and helps equalizing air pressure inside and outside the chamber. The protective cover serves to protect activated charcoal from water precipitation and has a small side hole to provide natural gas exchange between the atmosphere and the soil. This design and composition of the chamber allows to overcome numerous technical problems arising at radon accumulation:

a) the air pressure differences inside and outside the chamber are eliminated;
b) the convection of atmospheric air and soil gas is natural;

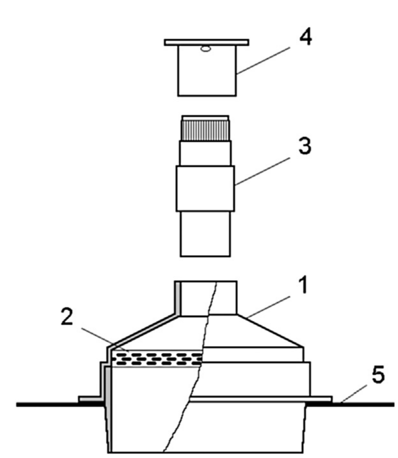

Fig. 1. The scheme of the accumulation open chamber with activated charcoal: 1 – accumulation chamber; 2 – activated charcoal layer (work layer) laid on a perforated bed; 3 – protective sorption column filled with activated charcoal (protective layer); 4 – protective cover with a hole; 5 – soil surface.