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Radon-222 in soil, water and building materials: Presentation of laboratory measurement methods in use at Risø

Claus E. Andersen
Risø National Laboratory, Build. NUK-125, DK-4000 Roskilde Denmark

Abstract

Three methods for measurements of radon-222 in soil, water and building materials are described, and sample results are given. The methods are used both in connection with the health problem of radon-222 in houses and with radon-222 as a tracer of environmental transport.

1 Introduction

Radon-222 constitutes a dominant part of the life-time radiation dose for most persons in the Nordic countries. This is, however, not the only reason why radon-222 is an interesting object of research: radon-222 is also an excellent tracer of certain transport processes in parts of the environment. At Risø, we are engaged in studies of both of these aspects. To this end we have adapted laboratory methods relating to radon measurements in soil, water and building materials. The purpose of this contribution is to describe three such methods, and to outline some results.

2 Emanation from soil samples

The emanation rate of soil is the number of radon atoms that (effectively) escape the soil grains into the pore system per kg dry mass per second. The emanation rate partly controls the soil-gas radon concentration. It is measured as follows: The sample is mildly disaggregated by forcing it through a brass plate with a 22 mm hole. The sample (typically about 300 g) is placed in a 6 L steel chamber. The chamber is flushed to near-zero radon concentration with about 30 L of aged nitrogen from a pressurized cylinder. Thereafter the chamber is closed, and radon starts to build up inside the chamber. Over the following days (or weeks) the chamber radon concentration is determined at selected times. Evacuated 200 mL scintillation cells are used for the purpose. An airbag with aged nitrogen is used to balance the pressure in the chamber after sampling. The scintillation cells are counted on a computer-controlled sample-changer with one photomultiplier tube. Samples are weighted before and after analysis. A moisture determination is also carried out. The analysis of the data includes corrections for dilution of the chamber because of sampling and pressure and temperature dependent transfer coefficients. Analysis of precision is carried out both for each chamber concentration determination (typically each cell is counted 3 to 4 times) and for the overall emanation-rate analysis (typically each analysis is based on 3 or more scintillation cell samples). The analysis-of-precision is a chi-squared test where the a priori uncertainty of the quantity of interest is compared with the experimental standard deviation. The final result of the analysis is reported on standardized computer-generated measurement sheets. Normally, batches of 12 samples are analyzed. In total, 480 analyses have been carried out. About 250 of these come from the Geological Survey of Denmark: To help understand differences in indoor radon potential for different geologies, samples from different

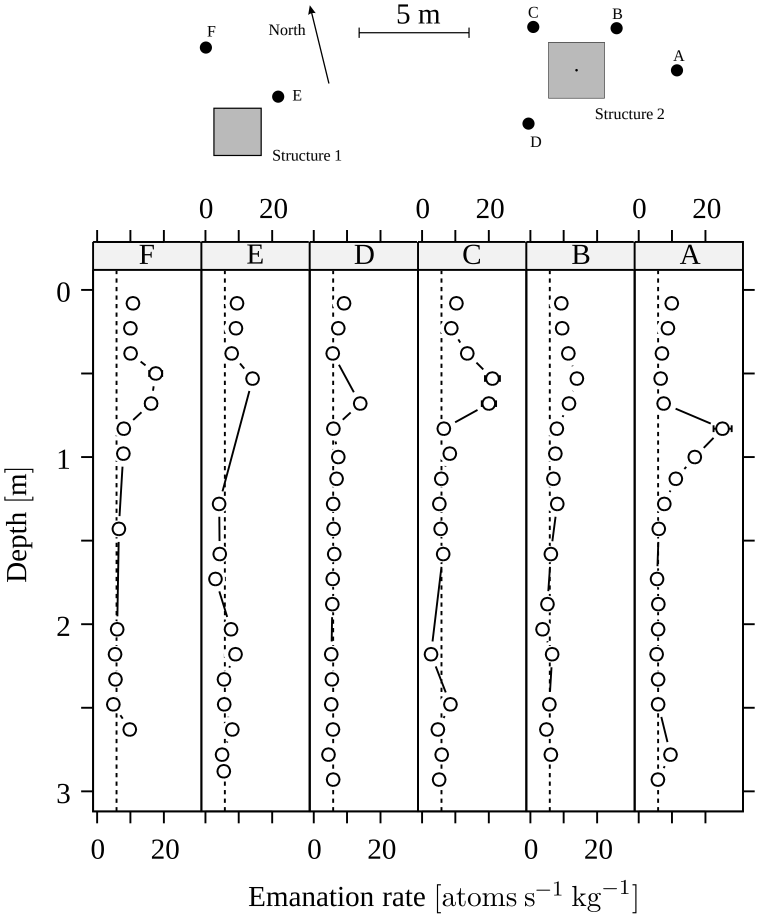

Fig. 1. Emanation rate results for six soil cores (named A to F) taken at the clayey till field site of Risø's radon test structures. The site is sketched above the graph. The dashed reference line at 5.8 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$ is the mean of all results below 1.4 m. Uncertainties (expressed as single standard deviations) are in most cases smaller than the plotted data points.

parts of Denmark have been analyzed. A small number of analyses have been done for engineering companies to help assess the risk for high indoor radon for larger construction works. About 200 emanation-rate determinations are from the site of Risø's radon test structures (the first structure, which is no longer in existence, is described in (Andersen, 1992)). Part of these analyses have been used to investigate the effect of disaggregation and moisture content (Andersen, 1998). Another purpose has been to provide soil parameters needed for the comparison of numerical models of radon transport in soil and entry into houses. Figure 1 shows 95 emanation rate results for six 3 m soil cores from the test structure site. Below 1.4 m depth, there is little variability with depth and from core to core. The pooled mean and standard deviation of these results amount to 5.8 and 1.4 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$, respectively (N=48). For the top (0–1.4 m) layer, the mean and standard deviation are 10.1 and 4.4 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$, respectively (N=47). It can be seen from the figure, that the emanation rate peaks in the layer between 0.5 to 0.85 m below the surface. The maximum value of 25 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$ occurs at 0.83 m depth for profile A. The high-emanation layer may result because the layer is more rich in radium than the other parts of the profile. Another possibility is that the fraction of emanation for the layer is high. The detailed mapping based on soil cores seems to allow for a much more clear understanding of the emanation-rate conditions of the site than previous measurements (cf. results given in (Andersen, 1992)).

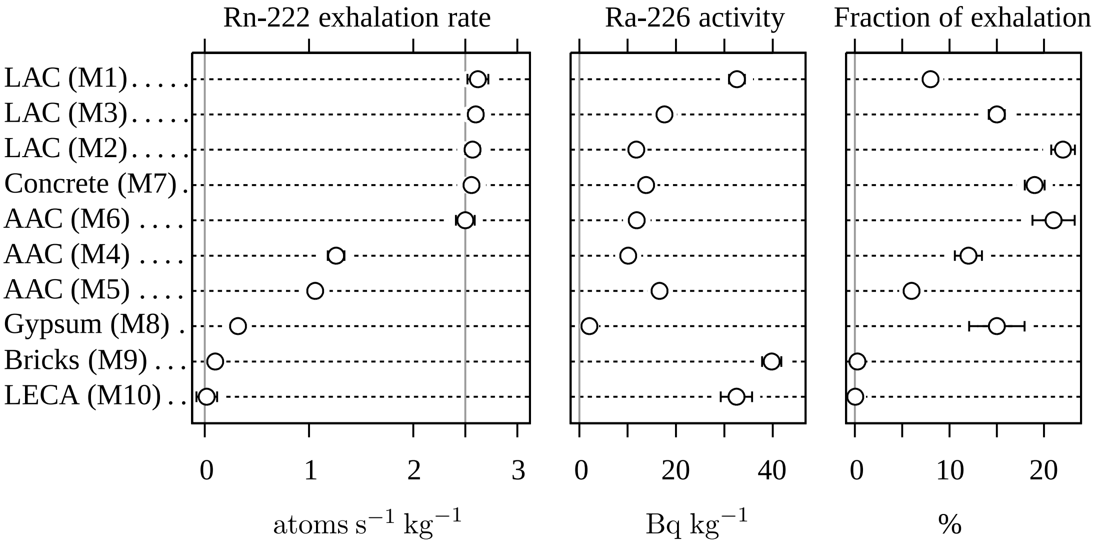

Fig. 2. Exhalation rate results. The meaning of abbreviations is given in the text. M1 means material 1 etc. The fraction of exhalation is the exhalation rate divided by the radium-226 concentration. Uncertainties (expressed as single standard deviations) are in many cases smaller than the plotted data points.

3 Exhalation from building materials

The mass-specific exhalation rate of radon-222 from a building material sample (such as a brick or a concrete slab) is the amount of radon-222 that escapes the sample per kg per second. Studies carried out by Jonassen, Ulbak and coworkers in the 1970'ies and 1980'ies showed that the radon-222 exhalation rate of ordinary Danish building materials is low. Special building materials with large radon-222 exhalation rates do however exist (at least in other countries). For this reason, it is of interest for Danish producers of building materials to be able to quantify this aspect of their products. Risø has therefore set up a method for exhalation rate measurements. It is a closed-chamber method. The sample (typically 30 x 30 x 5 cm^3) is placed in a 55 L stainless steel chamber together with a radon monitor that measures the radon-222 concentration every hour. Also temperature, humidity and pressure in the chamber is registered. The sample is conditioned for 24 h with a flow of aged nitrogen. The flow has a relative humidity of about 50 %. After conditioning, the chamber is closed and the radon concentration starts to build up. The measurement extends from 3 to 10 days. The sample is weighted before and after measurement. The method is documented in a report (Andersen, 1999) together with measurement results for 10 Danish building materials. All materials were found to have exhalation rates below 2.7 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$. The highest value were for ordinary concrete, lightweight aggregate concrete (LAC) and autoclaced areated concrete (AAC). Bricks, gypsum and lightweight expanded clay aggregate (LECA) had values below about 0.3 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$. Under consideration of the application of the mateials in a typical Danish single-family house, it was found that such materials cannot increase the indoor radon concentration by more than 10 $\mathrm{Bq\,m^{-3}}$. The Danish Institute for Radiation Hygiene (SIS) has measured the radium-226 concentration of the samples. The results are shown in Figure 2. The figure shows, for example, that some of the materials have a fraction of exhalation above 20 %. Other materials (such as bricks) have a very low fraction of exhalation. Although bricks have the largest radium concentration, very little radon exhale from the surface (the fraction of exhalation is less than 1 %).

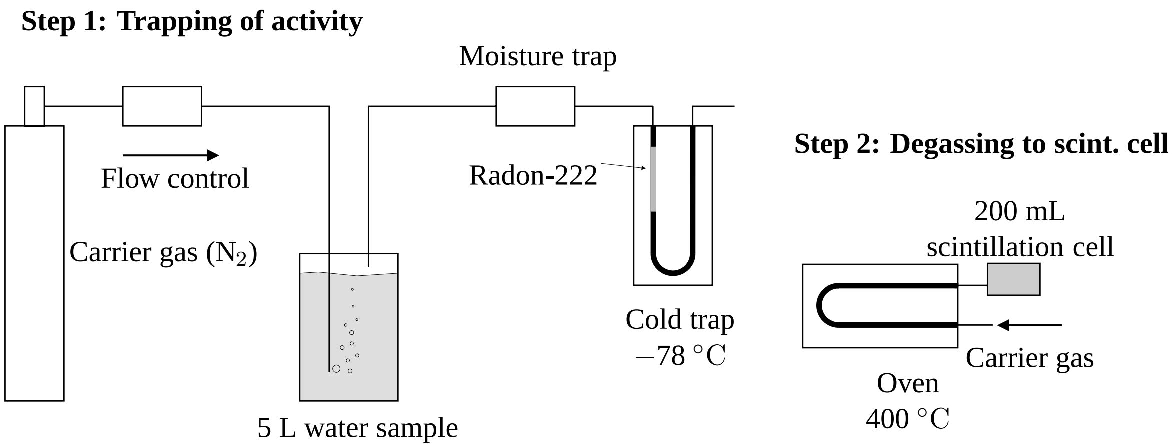

Fig. 3. Radon-222 degassing of seawater samples. The water sample is stripped for radon by flushing it with 30 L of nitrogen. The activity is collected on activated charcoal cooled to dry-ice temperature. In step 2, the trap is heated to 400 °C, and the activity is transferred to an evacuated scintillation cell.

4 Radon in seawater

Groundwater tends to have a high radon-222 concentration compared with seawater. Radon-222 can therefore be used to trace the sub-marine supply of ground water to the sea. In 1998 Risø engaged in an EU-project called sub-Gate which deals with submarine ground water fluxes and transport processes from methane rich coastal sedimentary environments. In this project, an area of Eckernförde Bay in the western Baltic Sea is investigated. In the area many so-called *pockmarks* exist, and ground water (containing radon) is believed to seep into the sea. Because of the relative close distance from the study area to Risø, a method has been adopted where samples are transported back to the laboratory for radon analysis. The method is sketched in Figure 3. It has been modified from that described by Mathieu *et al.* (1988). Cruises have been made in December 1998 and in July 1999. Preliminary results indicate that excess radon-222 exist close to the sea floor. In December 1998, near sea-floor radon-222 concentrations as high as 30 $\mathrm{mBq\,L^{-1}}$ were observed. In contrast, near sea-surface radon-222 concentration were only about 3 $\mathrm{mBq\,L^{-1}}$. The radium-226 concentration was found to be less variable. The average of 19 measurements was 3.3 $\mathrm{mBq\,L^{-1}}$.

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