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A RADON-EXHALATION MONITOR

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Radiation Protection Unit
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Abstract

A radon exhalation monitor has been constucted and calibrated. The monitor is based on the absorption of radon by active charcoal and the subsequent measurement of its γ activity by means of high resolution γ spectrometry. Exhalation rates may be determined with an accuracy of better than 20 % within 48 h after exposure. The detection limit is estimated on the basis of counting statistics to about 0.1 mBq/m^2/s.

submitted to Rad. Prot. Dosim.

1. Introduction

Radon exhalation from porous materials naturally occuring, like soil and rocks, as well as man-made, like mining wastes and building materials has been object of research activities since several decades [1,2,3]. In the case of exhalation from soil, the most commonly used method is to place a sample in a closed container and measure the growth of radon activity as a function of time. From this method it is to be expected that the obtained exhalation rate is a function of the sample size as well as its shape. Furthermore, the environmental conditions are drastically changed and may also falsify the result.
A simple method to measure ^{222}Rn exhalation from surfaces was introduced in ref. [4] based on the adsorption of radon by activated charcoal with succeeding $\gamma-$ spectrometry of radon decay products formed in the charcoal. The working principle of the presented radon-exhalation monitor is similar to that in ref. [4].

2. Design of the radon-exhalation monitor

The exhalation monitor consists of a funnel with a perpendicular raised border, which is placed upside down and partly pushed into the ground. The exhalation monitor covers an area $S_\epsilon = 0.126$ m^2. Under the funnel a canister filled with 245 g active charcoal is placed as shown in Fig. 1. In order to keep the radon long enough inside the funnel volume, the tube at the top is filled with glassfiber material (hatched area in Fig. 1). At the top of the tube a rain cover is mounted with some holes for sufficient air ventilation. A detailed sketch of the charcoal canister is given in Fig. 2. The active charcoal adsorbs the radon which exhales from the ground. The small opening at the top of the funnel prevents the build-up of a pressure difference between the air inside and outside the monitor. Although the active charcoal is covered by a layer of a drying medium (10 g silicagel), the competitive adsorption of vapour has to be taken into account and limits the exposure period to about 24 hours. In order to prevent from radon loss during transport the charcoal canisters are sealed with adhesive tape.
After exposure the adsorbed radon is measured by means of high-resolution $\gamma-$ spectrometry. The obtained signal is a measure of the exhalation rate from the ground. Before

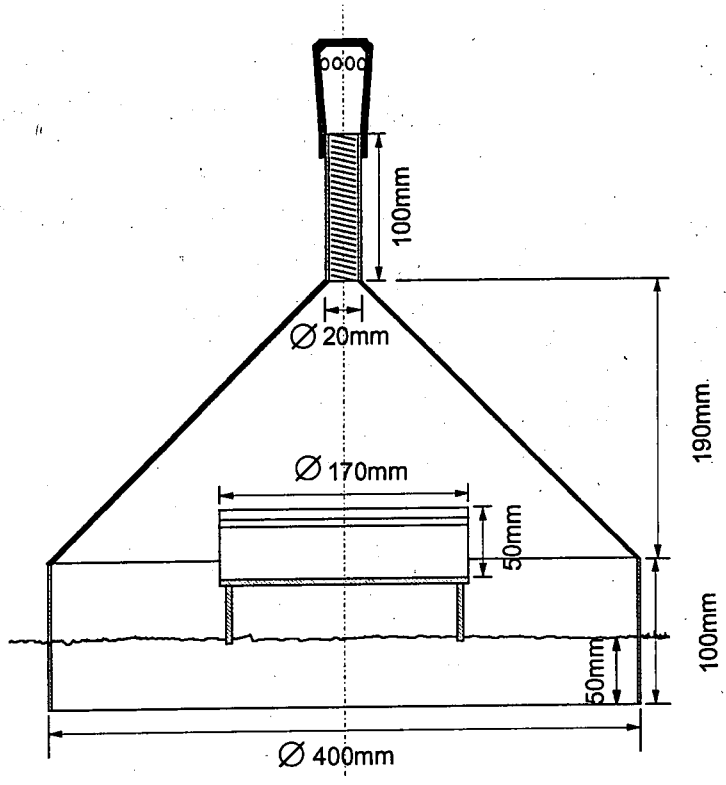

Fig. 1: Sketch of the exhalation monitor; the charcoal canister is placed on a stand to collect from the whole area covered by the funnel. The tube is filled with glassfiber material (hatched area) to keep the radon long enough inside the funnel. At the top of the funnel a rain cover is mounted.

exposure all radon and vapour has to be heated out of the charcoal at 130 °C for about 24 h.

3. Feasibility study and calibration

For the feasibility study the exhalation process is simulated by flushing a defined amount of radon through a vessel with approximately the same volume as the funnel, which contains the charcoal canister. A sketch of the setup is shown in Fig. 3. Cycle (1) is used to flush

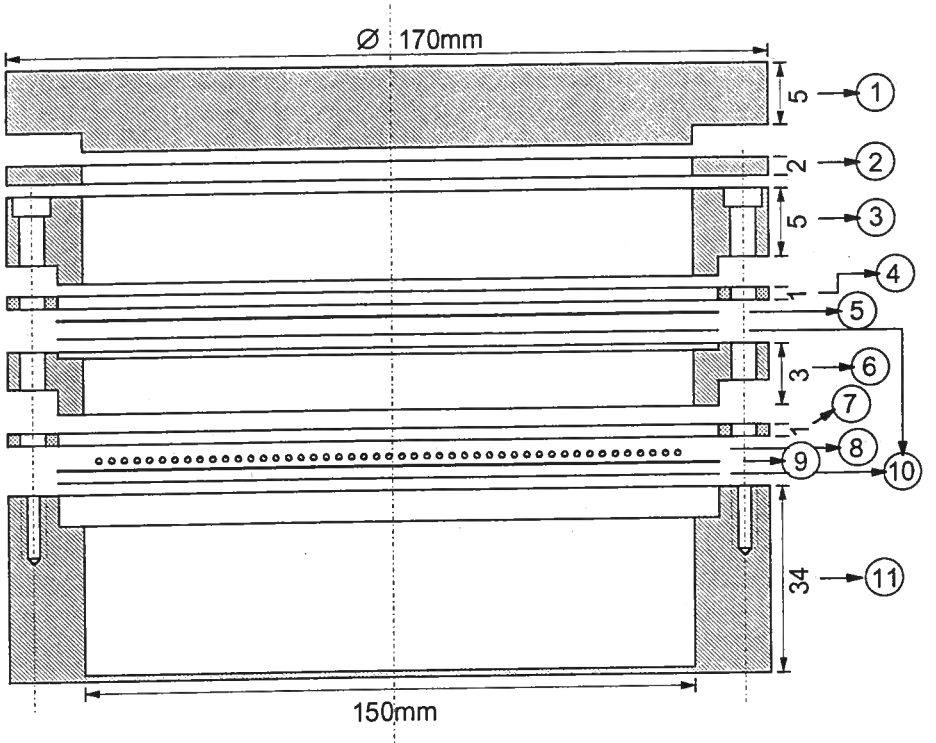

Fig. 2: Detailed sketch of the charcoal canister used for radon exhalation monitoring (all measures in mm). The canister is made of aluminium, and the top is made of stainless steel. (2), (4), (7) : rubber seals; (5), (9), (10) : screens with different mesh size to cover the activated charcoal and to separate it from the drying medium (8, silicagel); (3), (6) : o-rings to fix the construction; (11) pot which containes 245 g activated charcoal.

the air inside V_0 over a radon source. The actual radon activity concentration in V_0 is monitored continuously with a Lucas-type monitor [5,6] with an integration period of 1 h. Afterwards the air is circulated through cycle (2) over the charcoal canister in the vessel (V). The air flow is measured by a rotameter.

The radon activity adsorbed during exposure is then assessed by measuring the $\gamma-$ activity of the radon daughter ^{214}Bi at an energy $E_\gamma = 609.3$ keV. The $\gamma-$ spectrometer consists of a high purity germanium detector and is calibrated with a ^{134}Cs source, which provides a $\gamma-$ line at $E_\gamma = 604.7$ keV close to that of ^{214}Bi. The ^{134}Cs activity is distributed in a homogeneous solution in the respective geometry and within half the height of the charcoal

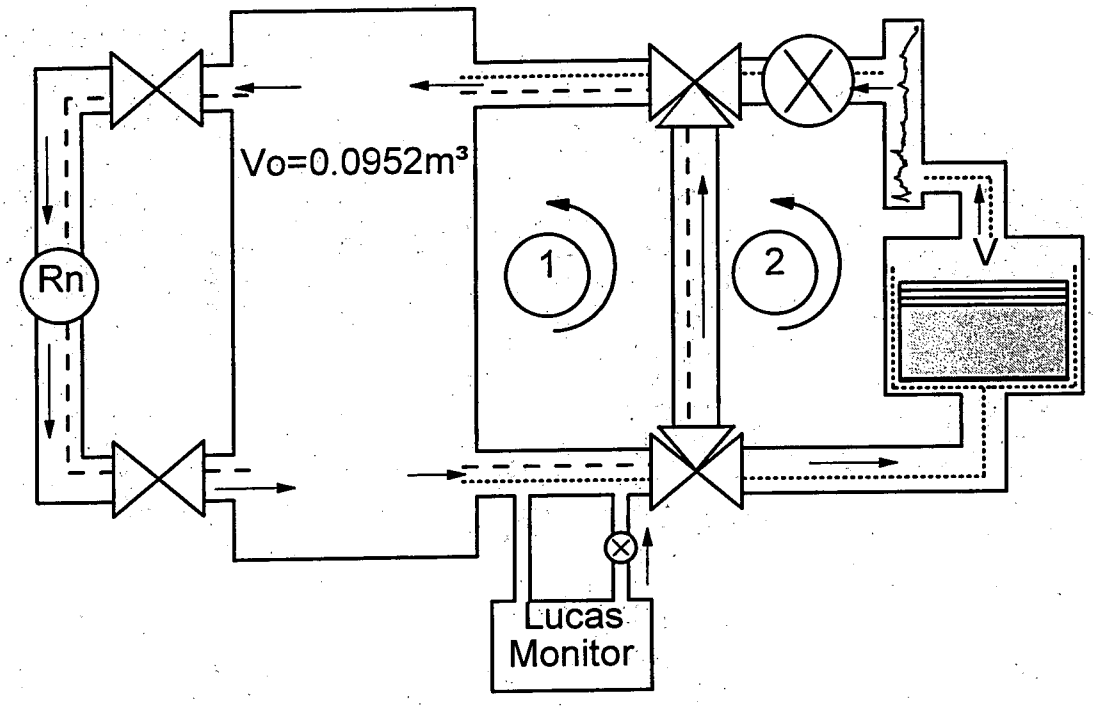

Fig. 3: Setup used for the calibration of the exhalation monitor. Cycle (1) is used to flush the air inside V_0 over a radon source. The radon concentration is continuously monitored with a Lucas-type monitor. Then, the air is circulated through cycle (2) over the charcoal canister which is placed in the vessel (V). The air-flow is measured by a rotameter.

canister. In order to mimic the influence of radon diffusion inside the charcoal canister, the efficiency of the $\gamma-$ spectrometer is determined for different distances between the source and the detector end-cap. Taking into account the respective photon branching ratio for the caesium as well as for the bismuth line the efficiency of the $\gamma-$ spectrometer ranges from 0.6 to 0.7 %.

In different runs two different charcoal canisters were calibrated at similar radon exposures. In both cases the exposure period t_E was 16 h. In Fig. 4 the respective radon activity c_{Rn} during exposure is shown. The lines are a guide to the eye. Afterwards the exposed charcoal canisters were placed in the $\gamma-$ spectrometer in order to measure the

^{214}Bi activity. The measured peak area at E_γ = 609.3 keV (N_{exp}) was then compared with the expectation value (N_{calc}) derived from the amount of adsorbed radon. Each canister was measured several times over a period of about four days. The ratio N_{exp}/N_{calc} as a function of time after exposure is shown in Fig. 5. The symbols and lines correspond to those in Fig. 4.

From the data in Fig. 4 an adsorption half life can be estimated of about 5 hours. This is the time needed by the charcoal canister to adsorb half of the radon out of the volume of about 110 l (V_0 + V). The air volume of the funnel is only 10 l resulting in an adsorption half life of less than 1 h. Thus, with exposure periods of more than 10 hours all exhaled radon may assumed to be adsorbed on the charcoal.

As it is obvious from the data in Fig. 5, there is agreement between the amount of adsorbed radon and the measured bismut activity indicated by a ratio almost equal to one. We see, that during the first 20 h radon diffusion from the surface region into the canister takes place leading to a slightly increasing ratio. The margin of this increase is compatible with the change of the spectrometer's efficiency estimated above for the effect of radon diffusion. After about 48 h leakage of radon from the canister becomes the dominant process, and the ratio decreases again, gradually. Thus, if the analysis of a charcoal pot is performed within 48 h after exposure, the adsorbed radon activity may be determined within a systematic uncertainty of $\pm$ 10 %.

A second important observation from Fig. 5 is the feature of reproducibility, i. e. the tested canisters behave similar with respect to both diffusion and leakage.

4. First in-situ measurements

For measurements on radon exhalating ground, the measured bismut signal has to be converted to a radon exhalation rate (ϵ_{Rn}), i. e. a radon activity release per unit area and unit time expressed in (m)Bq/m^2/s. Since the exposure period t_E is much longer than the half lives of the decay products, radioactive equilibrium amongst them may be assumed for conversion. Thus, the adsorbed radon activity A_0 at the end of exposure can be calculated from the number of bismut decays N_{exp} measured by means of $\gamma-$ spectrometry between t and t + Δt to a good approximation by

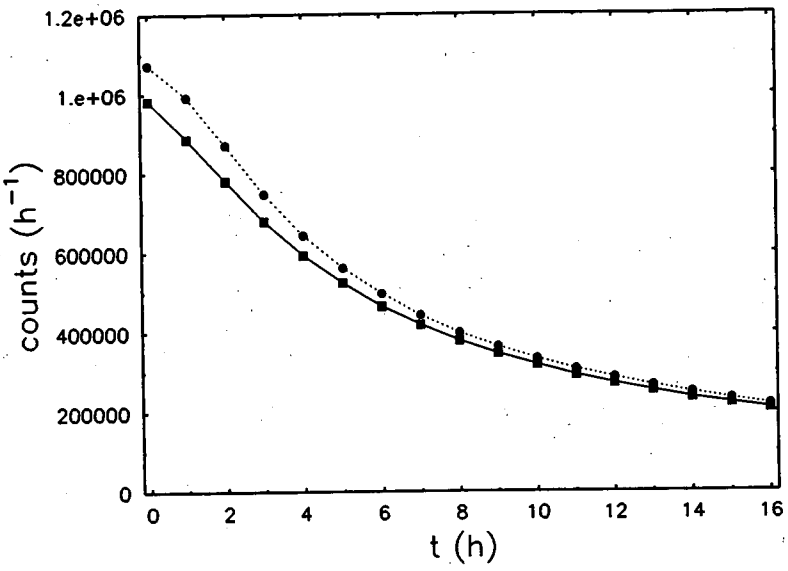

Fig. 4: Radon activity during the exposure of two different charcoal canisters (the lines are a guide to the eye).

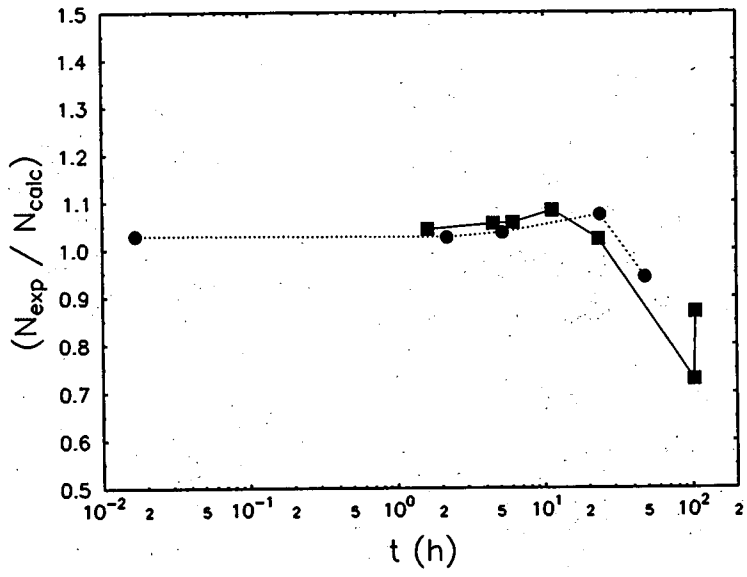

Fig. 5: Ratio between the measured and calculated number of counts, N_{exp} and N_{calc}, respectively, as a function of time after exposure. The symbols were taken according to Fig. 4.

$$A_0 = (\lambda_{Rn}/b_\gamma)\, N_{exp} \exp(\lambda_{Rn} t)/(1 - \exp(-\lambda_{Rn}\Delta t))\,, \qquad (1)$$

where b_γ is the photon branching ratio, and N_{exp} is already corrected for the efficiency of the $\gamma-$ spectrometer. From A_0 the radon exhalation rate ϵ_{Rn} can be calculated from the solution to the differential equation for the radon activity growth inside the active charcoal, due to continuous absorption of radon from the air inside the funnel volume :

$$\frac{dA}{dt} = -\lambda_{Rn} A + \epsilon_{Rn} S_\epsilon\,, \qquad (2)$$

where S_ϵ denotes the exhalation area covered by the funnel. Finally, the radon exhalation rate is calculated according to

$$\epsilon_{Rn} = (\lambda_{Rn}/S_\epsilon)\, A_0/(1 - \exp(-\lambda_{Rn} t_E))\,. \qquad (3)$$

First in-situ measurements have been performed on the SCK•CEN domein. The soil consists of sand with a low specific radium activity $A_{Ra} = 13$ Bq/kg [7] (world average value $\langle A_{Ra}\rangle = 25$ Bq/kg [8]). The ground-water level is about 1 m below the surface. According to ref. [9] a radon exhalation rate from this soil may be estimated from

$$\epsilon_{Rn} = \langle\varepsilon\rangle\, A_{Ra}\, \rho\, R_B \lambda_{Rn}\quad, \qquad (4)$$

with the radon emanation power $\langle\varepsilon\rangle \approx 0.1$, the radon decay constant $\lambda_{Rn} = 2.1 \times 10^{-6}$ s^{-1}, the soil density $\rho \approx 2 \times 10^3$ kg/m^3 and a radon diffusion diffusion length $R_B \approx 1.5$ m leading to an estimated value for $\epsilon_{Rn} = 8$ mBq/m^2/s with an uncertainty of about 30 %.

Four different exhalation monitors were placed together on an area of about 2 m^2. The monitoring period was 16 h. After exposure all charcoal canisters were analysed by means of $\gamma-$ spectrometry at least for two times, directly after exposure ($t < 10$ h) and again about 24 h later. The results are shown in Fig. 6, where different symbols refer to different charcoal canisters. It is satisfying to see the consistency amongst the obtained exhalation rates. The average exhalation rate is 5.8 mBq/m^2/s with an uncertainty less than 9 % at

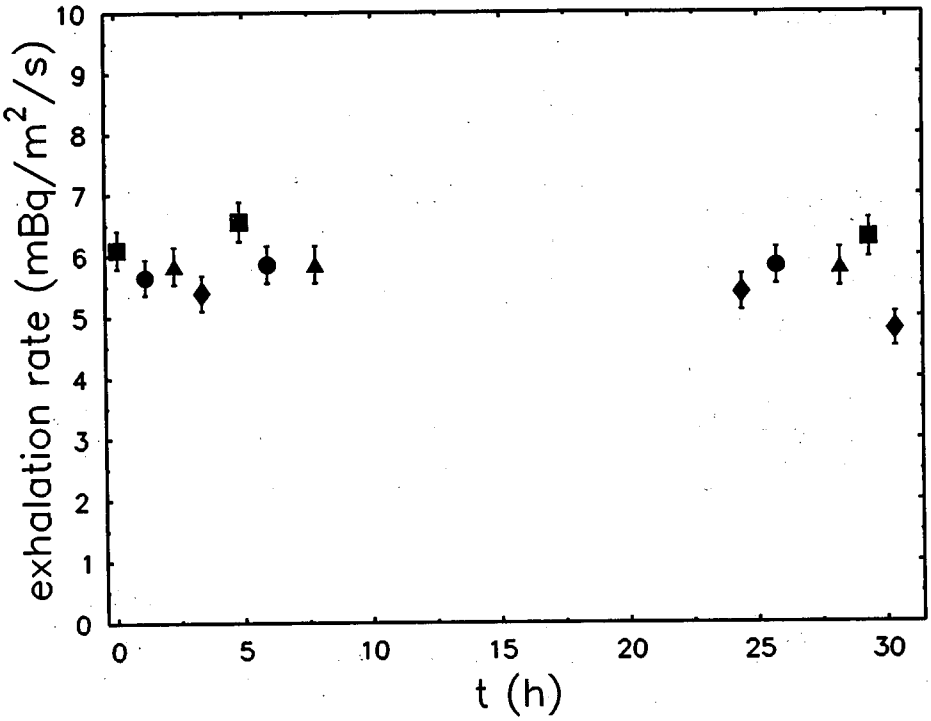

Fig. 6: Results from in-situ measurements with radon exhalation monitors presented in this text. Different symbols indicate different monitors

one standard deviation. This value is compatible with the previous estimate and on typical exhalation rates from soil ranging from 1 to 50 mBq/m^2/s as shown in ref. [9].

5. Summary

A radon exhalation monitor has been constructed based on the adsorption of radon by active charcoal. The feasibility study shows that the measured activity is a direct measure for radon exhalation rates with an accuracy of about 20 % within 48 h after exposure. The minimum detectable exhalation rate is estimated on the basis of counting statistics at 0.1 mBq/m^2/s for exposure periods of about 16 h and measurement periods of about 2 h. With first in-situ measurements the expexted accuracy and reliability of the presented

radon exhalation monitor could be confirmed.

References

[1] Kovach E. M., *Meteorological Influences upon the Radon-Content of Soil Gas*, Trans. Am. Geoph. Union 26 (1945) 241

[2] Jonassen N. and J. P. McLaughlin, *Exhalation of Radon-222 from Building Materials and Walls*, in : The Natural Environment III, Vol. 2, Sympo. Proc. (Edited by T. F. Gesell and W. M. Lowder) Houston, TX, 23.-28. April 1978, NTIS, CONF-780422 (1980) 1211

[3] Keller G. and M. Schütz, *Radon Exhalation from the Soil*, Rad. Prot. Dosim. 24 (1988) 43:46

[4] Countess R. J., ^{222}*Rn Flux Measurement with a charcoal canister*, Health Phys. 31 (1976) 455:456

[5] Lucas H. F., *Improved low-level alpha-scintillation counter for radon*, Rev. Sci. Inst. 28 (1957) 680

[6] Pylon model AB-5, *Instruction manual A900024*, Pylon Electronics Inc. (1985) rev. 5. 06. 1993

[7] Deworm J. P., W. Slegers, J. Gillard, J. M. Flemal and J. P. Culot, *Survey of the natural radiation of Belgian territory as determined by different methods*, Rad. Prot. Dosim. 24 (1988) 347

[8] UNSCEAR, *Sources, effects and risks of ionizing radiation*, Report on the general assembly, with annexes, United Nations Sales Publication E.88.IX.7 (1988) New York

[9] Porstendörfer J., Fifth International Symposium on the Natural Radiation Environment (tutorial sessions), edited by the Commission of the European Communities, EUR 14411 EN (1993) 69 unpublished