SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ÉTUDE DE L'ÉNERGIE NUCLÉAIRE

RADON IN THE INDOOR ENVIRONMENT (A STATUS REPORT)

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Radiation Protection Unit
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BLG-679

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Abstract

In the following a summary of investigations on *Radon in the Indoor Environment* is given, which were performed within the young scientists programme of the SCK•CEN. In the frame of a two-years contract a new personal radon dosemeter with a high efficiency was designed and calibrated. Furthermore, a so-called bronchial dosemeter was built to assess the deposition characteristics of short-lived radon decay products in the human respiratory tract. The calibration was completed and first successful measurements were performed. Finally, the $\alpha-$ activity of ^{210}Po trapped in spongy materials was investigated for the first time as a retrospective radon monitor. A very sensitive and accurate tool for the assessment of radon exposures in dwellings has been developed, which accuracy is higher by more than one order of magnitude compared to other common techniques based on the analysis of the ^{210}Po-activity in surface layers.

Introduction

Half of the human exposure to ionizing radiation comes from radon progeny in the indoor environment. Different surveys showed, that high radon concentrations are not limited to mines and industrial facilities but exist also in domestic environments due to radon exhalation from radium in the ground. The very large variation in radon concentrations results in a small group receiving doses higher than the limit for workers. A correlation between radon exposures and an increased lung-cancer risk was found in epidemiological studies of miners. The radiation dose to the lung is due to the inhalation of the airborne short-lived radon decay products ^{218}Po, ^{214}Pb and ^{214}Bi(^{214}Po), which deposit in the respiratory tract. In the indoor environment this deposition strongly depend on the attachment rate of the freshly formed decay products to aerosol particles and on the plate-out rate to indoor surfaces. Although it is known that the unattached fraction of the decay products is an important contributer to the radiation dose, the assessment of this fraction on the basis of wire-screen methods were only partially successful. Furthermore, since it is known, that lung-cancer is caused by radon exposures over a long time period, for the estimate of the lung-cancer risk induced by radon as well as for epidemiological studies a precise retrospective assessment of long-term radon exposures in dwellings is essential.

A few years ago activities have been started in the radiation protection research unit of the Belgian Nuclear Research Center SCK•CEN in the field of radon monitoring. For monitoring purposes flow-through Lucas-cells are used. Short-term measurements (1 d) are performed with active-charcoal detectors. Radon monitoring on a time-scale of typically 6 month is based on so-called Karlsruhe-type polycarbonate track-etch detectors. Additionally, the radon-group carries out contract reseach for industrial firms dealing with radon exhalation from building material as well as with permeability studies on plastic foils.
In the frame of the young scientists programme new research activities were started at SCK•CEN in order to explore new measurement techniques for radon and radon progeny monitoring. The whole radon research programme consists of three major projects :

- Monitoring of individual radon exposures. Design and calibration of a personal radon dosemeter, which is capable to monitor individual exposures down to average radon concentrations typical for Belgian dwellings within an integration period of one working month (= 170 h). Further recommendations were to establish an easy dosemeter's handling as well as a high cost effectiveness.

- Investigation of the deposition characteristics of the short-lived radon daughters in the human respiratory tract by means of the 'bronchial dosemeter'.
- Retrospective assessment of radon exposure in dwellings by investigating the $\alpha-$ activity of ^{210}Po trapped in spongy materials.

In the following a summary of the achieved results for each project is given. For more details it is referred to different publications listed at the end of this report. A brief outlook for further activities is added to the report.

Achievements

Design and calibration of a personal radon dosemeter

For personal monitoring, in general, passive radon detectors are used based on the counting of etched tracks produced by $\alpha-$ particles stopped in plastic materials. At SCK•CEN a closed-type dosemeter has been considered. This type avoids any influence of humidity, dust and accidential surface damage on the response of the detector material. The dosemeter is based on a small box made of plastic, which is commercially available. The detector is placed at the back side of the dosemeter as shown in Fig. 1. As detector material CR-39 (polyallyl diglycol carbonate) track-etch detectors are used, which is sensitive to the complete range of $\alpha-$ particle energies from the decay of radon and its daughters, and which is widely used in different fields of nuclear physics for charged particle detection. Since the diameter of $\alpha-$ particle tracks in CR-39 after etching are only of the order of μm the track-counting had to be performed at a magnification of about 200. In order to keep the systematic uncertainty of the read-out of the dosemeter as low as possible, the $\alpha-$ particle track-counting procedure was based on the image-analysis technique performed on a personal computer. A typical image of etched $\alpha-$ particle tracks in CR-39 material is shown in Fig. 2. Details on the detector handling, automatized track-counting and the etching conditions applied to the CR-39 detector material are given in Publ. 4 listed at the and of this report.

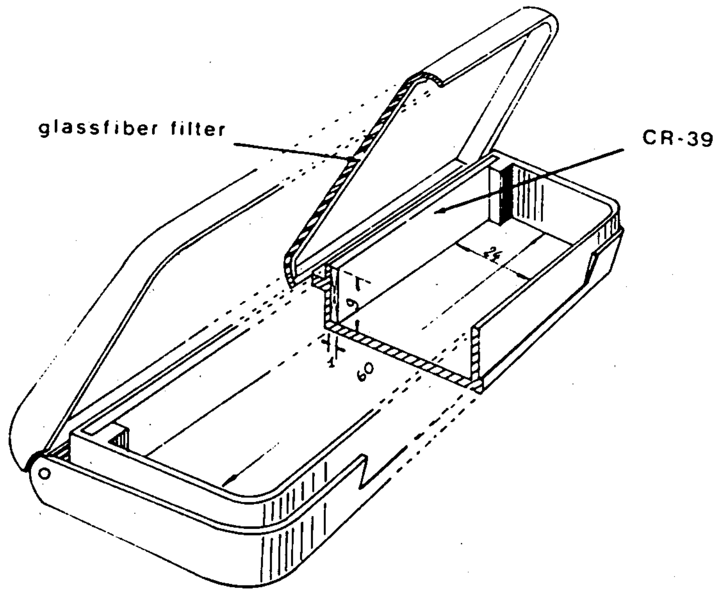

Fig. 1: Schematic representation of the SCK•CEN personal radon dosemeter (all measures in mm) The CR-39 detector is placed on the backside of the dosemeter

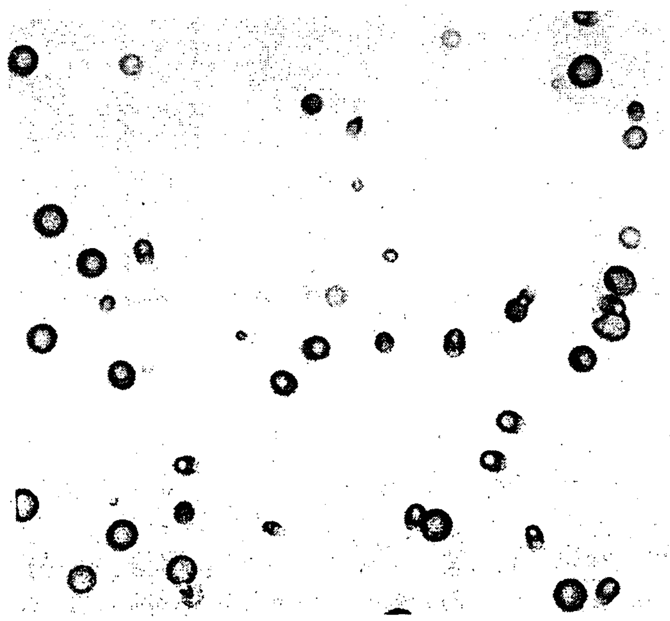

Fig. 2: Example of etched $\alpha-$ particle tracks in CR-39 material. The size of the tracks is of the order of μm and depends on the $\alpha-$ particle energy. The shape of a track depends on the incident angle of the $\alpha-$ particle on the detector's surface.

Design and calibration of the new personal dosemeter has been completed in early 1994. The efficiency ϵ_{PRD} after automatised track-counting is

$$\epsilon_{PRD} = 1.43 \pm 0.15\,\text{tracks/cm}^2/(\text{kBq/m}^3\text{h}) \quad , \tag{1}$$

which is three times higher than that of the standard Karlsruhe-type track-etch detector. The detection limit at 20 % uncertainty within the required exposure period of 170 h is 200 Bq/m^3. With an additional effort concerning e. g. the cleaning procedure applied to the detector material prior to radon exposure as well as the cutting technique of the CR-39 sheets the dosemeter's detection limit could be decreased by 50 %. Final results have been reported to the Belgian Ministry for Scientific Research.

The bronchial dosemeter

Basically, it is assumed that the deposition characteristics of the human respiratory tract may be simulated by different numbers of wire-screens. Their respective sizes and numbers are determined by comparing models of the respiratory tract with results from the screen-penetration theory. After collecting the airborne activity on membrane filters, the $\alpha-$ activity of ^{218}Po and ^{214}Po is measured for at least two periods, from which the radon-daughter concentrations can be calculated. Thus, the bronchial dosemeter consists of a sampling section in order to simulate the inhalation and deposition process during breathing and of a counting section to measure the $\alpha-$ activity due to the collected radon decay products on the filters. The sampling section consisted of three independent sampling channels with different numbers of screens in order to collect the total airborne and the penetrated activities through the nasal cavity as well as through the bronchial tree. A prototype of this so-called bronchial dosemeter has already been set up in 1992. During 1994 the bronchial dosemeter has been technically improved and recalibrated. The efficiency of each of the three sampling channels have been optimized to about 15 %. The efficiencies of all sampling channels were obtained from six different calibration runs with relative uncertainties less than 1.1 % (see Tab. 1). For details on the calibration procedure see Publ. 6 of the attached list.
First test measurements have already been performed during November 1994. The results indicate the ability of the 'bronchial dosemeter' to individually assess the fractional deposition of the different short-lived radon-daughters in the nasal cavity as well as in the bronchial tree.

Table 1: Efficiencies of the three sampling channels of the 'bronchial dosemeter' obtained from six measurements at a distance of 0.7 cm from the detector.

head(1)	head(2)	head(3)
0.1492 ± 0.0016	0.1620 ± 0.0017	0.1517 ± 0.0010

As an example the fractional depositions in the nose (f_n) and in the bronchial tree (f_b) of the short-lived radon daughters are shown in Fig. 3. Further measurements with the 'bronchial dosemeter' are necessary to study the influence of different aerosol conditions, which are characteristic in the domestic environment, on the deposition characterisitics.

Retrospective assessment of radon exposure

A new retrospective monitoring technique for radon exposure has been developed using volume traps. The goal was to measure the activity due to the $\alpha-$ decay of the radon progeny ^{210}Po, which is the granddaughter of ^{210}Pb ($T_{1/2}^{Pb} = 22$ y), deposited inside spongy materials as mattresses and cushions. The exposure of recently produced polyester material was performed during 1993. After approaching radioactive equilibrium the samples were ready for analysis in 1994. The complete analysis consists of different steps. First, the deposited ^{210}Po has to be separated chemically. The efficiency of this procedure is individually monitored for each sample by adding a radioactive tracer material (^{208}Po). A frequency distribution of the chemical efficiency ϵ_c is shown in Fig. 4. During our investigations a mean value of $\epsilon_c \approx 0.6$ was achieved. The small peak at low efficiencies around 0.12 is due to the initial use of less concentrated nitrogen acid during separation. At the end of the separation the Polonium autodeposits on a silver-plate, which finally is analysed by means of $\alpha-$ spectrometry.
Polyester samples with four different densities have been investigated. In Fig. 5 the measured specific sample activity a_s (mBq/g) is shown as a function of the radon exposure. Measurements on unexposed samples for background determination are included. Different symbols refer to different sample densities ρ_s according to Tab. 2. All data show a perfectly linear dependence on the radon exposure indicated by the different lines. The respective parameters of a linear regression are given in Tab. 2. These results indicate that this new technique is better by more than one order of magnitude compared to other common methods based on the measurement of ^{210}Po implanted in glass surfaces.

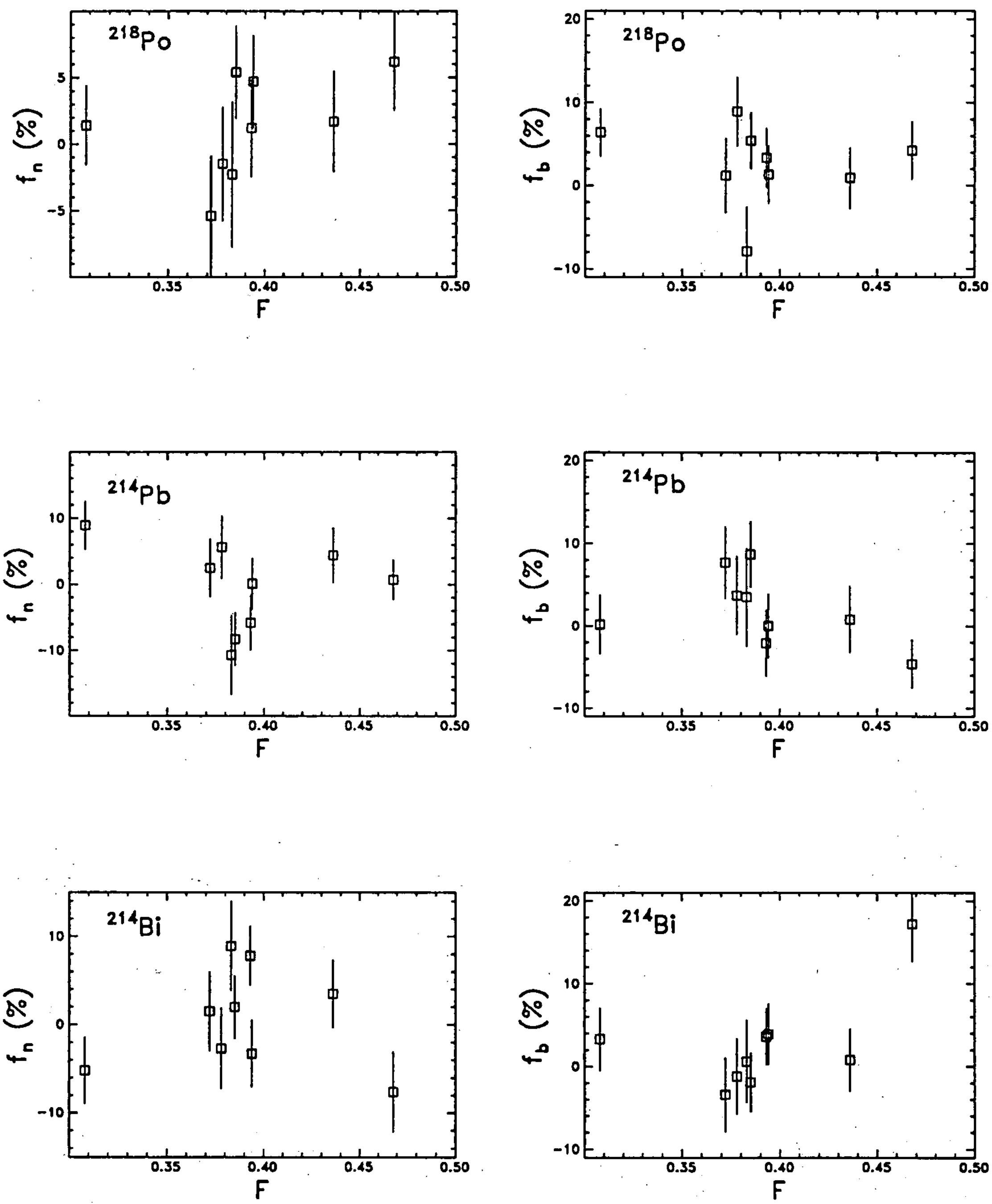

Fig. 3: Fractional deposition of the short-lived radon decay products ^{218}Po, ^{214}Pb and ^{214}Bi as a function of the equilibrium factor F. The left part shows the fraction of decay products deposited in the nasal cavity (f_n), and the right part shows the fractional deposition in the bronchial tree (f_b).

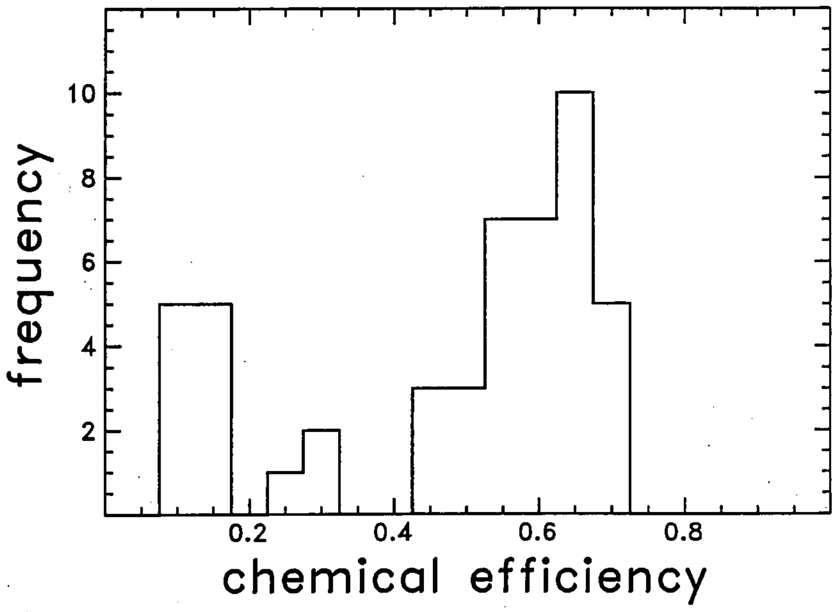

Fig. 4: Frequency distribution of the efficiency of the chemical separation of ^{210}Po deposited in polyester material.

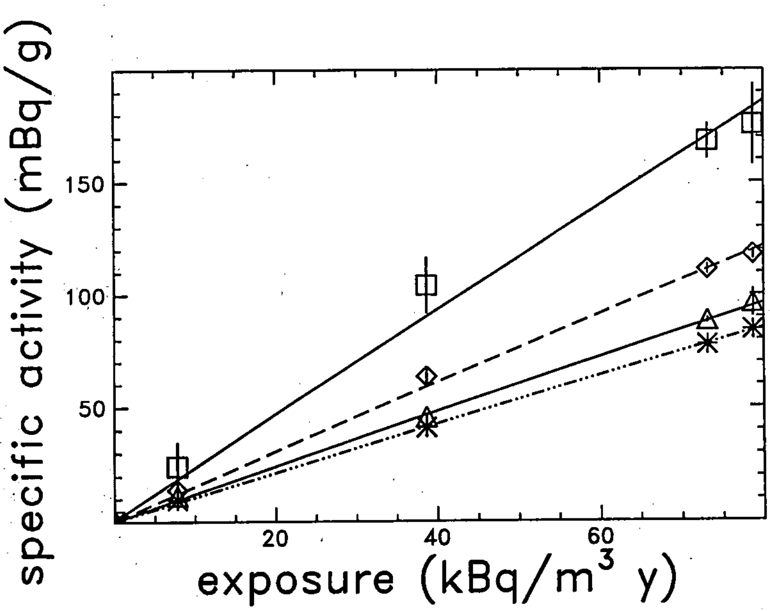

Fig. 5: Specific sample activity a_s as a function of the radon exposure. Different symbols refer to different sample densities. The order is according to Tab. 2. The lines indicate the respective fit to the experimental data.

Table 2: Sets of linear coefficients fitted to the specific sample activities a_s as a function of the exposure E.

sample id#	density ρ_s (kg/m^3)	a_0 (mBq/g)	a_1 (mBq/g)/(kBq/m^3 y)
1	20.3	0.59 ± 0.97	2.32 ± 0.08
2	29.3	0.38 ± 0.48	1.517 ± 0.024
3	40.3	0.23 ± 0.28	1.20 ± 0.01
4	44.1	0.15 ± 0.12	1.064 ± 0.009

In a next step the data have further been reduced by dividing them by the respective sample density ρ_s. This volume activity a_V expressed in mBq/cm^3 should be independent of ρ_s and furthermore, the conversion factor $a_{V(1)}$ can be directly derived from the number of deposited ^{210}Pb-atoms per unit exposure and the corresponding decay constant λ leading to

$$a_{V(1)} = 0.0455\,\text{mBq/cm}^3/(\text{kBq/m}^3\text{y}) \quad . \tag{2}$$

In order to compare this value with the experimental data a correction factor k_f for the effective free air volume inside the sample material has to be applied to the data, which is defined by

$$k_f = (\rho_m - \rho_s)/(\rho_m - \rho_l) \quad , \tag{3}$$

where ρ_m and ρ_l are the density of the material (polyester : $\rho_m = 1.05$ g/cm^3) and air ($\rho_l = 0.00125$ g/cm^3), respectively. Thus, the conversion factor has to be calculated by

$$a_{V(1)} = \rho_s\, a_1/k_f \quad . \tag{4}$$

Here an average value $\overline{k_f} = 0.969$ has been applied to the data. In Fig. 6 the volume activity (symbols) is shown as a function of the radon exposure together with the corresponding linear fit (full line). The fitted slope, i. e. the conversion factor,

$$a_{V(1)} = 0.0481 \pm 0.0009\,\text{mBq/cm}^3/(\text{kBq/m}^3\text{y}) \quad , \tag{5}$$

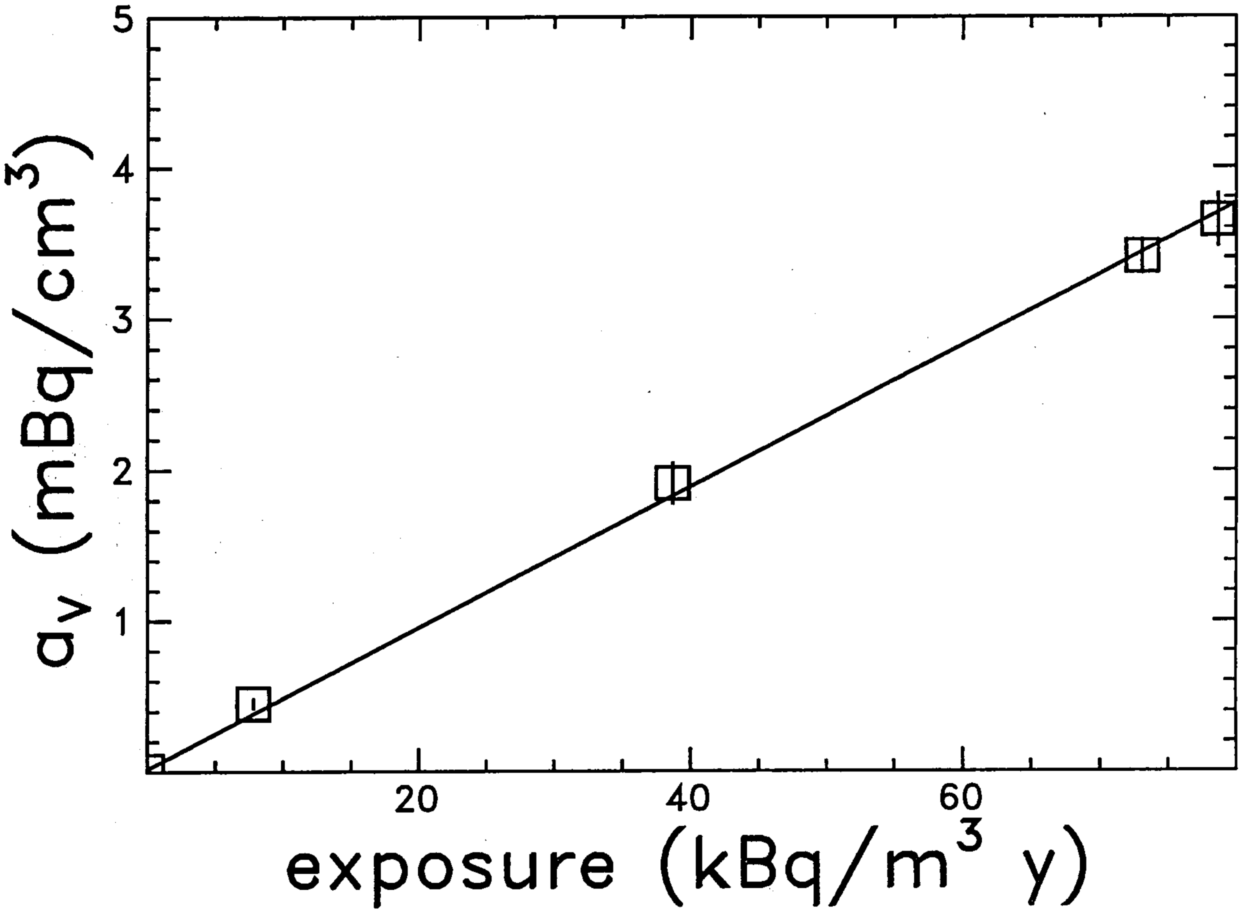

Fig. 6: Volume activity a_V as a function of the radon exposure. The full line gives the result of a linear fit to the data. The relative uncertainty on the fitted slope is 1.9 %.

which is in very good agreement with the expected value from eq. (2). Therefore, the new retrospective radon monitor is not only very accurate as well as very sensitive, but applicable to different types of volume-trap materials.

Future 'radonactivities' at SCK•CEN

Besides the existing experience in radon monitoring SCK•CEN actively contributed to the accomplishment of scientific problems in radon progeny assessment and risk evaluation methods within the last two years. Within an international framework under the auspices of the Commission of the European Communities (CEC) collaborations with several institutes from 6 countries could be established. This will probably result in a further participation of SCK•CEN in the next Radiation Protection Programme of the CEC.

The future research activities will in first instance be concentrated on two projects. Firstly, the new retrospective radon monitoring technique will be tested by analysing samples collected from houses with known radon concentrations. Secondly, a radon reference chamber will be set up at SCK•CEN. This reference chamber will serve for well controlled athmospheric conditions throughout the comprehensive investigations on the dynamical characteristics of the short-lived radon decay products in indoor air. The design and construction of this radon chamber will take place within an SCK•CEN research and development programme over a five years period.

The reference radon chamber will have a volume of about 5 m^3 and has to be air-tight. The ventilation rate will be controlled, and aerosol production and monitoring facilities will be installed. It will be possible to measure the unattached and aerosol-attached radon daughters in the size range of ions and clusters ($\sim$ 1 nm) up to larger aerosol particles (several μm). The whole project may roughly divided into three phases :

- The first phase deals with the construction and built-up of the radon reference chamber and is supposed to take about 2 years. This period also includes a detailed study of the necessary equipment for aerosol production and data acquisition.
- The second phase covering the 3rd and 4th year is related to the equipment installation and the setup of the online multi-parameter monitoring system. On particular will be a detailed investigation of specific aerosol sources. Already from the 3rd year on radon-related research and monitoring services will be possible under reference conditions.
- In the third phase, i. e. during the 4th and 5th year a detailed study of the physical and chemical interaction (particle growth, cluster formation, plate-out rates) of unattached radon progeny with trace gases and other aerosol particles as well as the deposition characteristics in the respiratory tract will be performed with the SCK•CEN bronchial dosemeter.

Under these reference conditions the SCK•CEN will be able to accurately simulate the deposition of radon progeny in the human respiratory tract. On that basis in conjunction with measurements in realistic domestic environments it will be possible to significantly improve the determination of the lung dose leading to an improved exposure-risk model.

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Scientific partners

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Georg-August Universität (Göttingen, Germany)
Université de Bretagne Occidentiale (France)
Universidad Politéchnica de Catalunya (Barcelona, Spain)
Institutionen för Radiofysik (Lund, Sweden)
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University of College Dublin (Ireland)

List of relevant publications and contributions to international conferences

1. *Calibration of a Polycarbonate Track-etch Detector*
   H. Vanmarcke, A. Toye, S. Oberstedt, Posterpresentation at the First International Workshop on Indoor Radon Remidial Action 27.06 - 2.07.1993, Rimini (Italy), *and* Rad. Prot. Dosim. Vol. 56, 1-4 (1994) 239

2. *Recent Investigations on Indoor Radon at SCK-CEN Mol*
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Annales de l'Association belge de Radioprotection, Vol 19, 1-2 (1994) 255

3. *Retrospective Assessment of Radon Exposure in Dwellings by Investigating Spongy Materials*
S. Oberstedt and H. Vanmarcke, Verhandlungen der DPG Frühjahrstagung Hamburg, März 1994

4. *Personal Radon Dosemeter*
S. Oberstedt and H. Vanmarcke, SCK•CEN Internal Report BLG 649, March 1994

5. *Volume Traps - A New Retrospective Radon Monitor*
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6. *The Bronchial Dosemeter*
S. Oberstedt, H. Vanmarcke, SCK•CEN Internal Report BLG 667, November 1994, *and* sent for publication

7. *Volume Traps as Retrospective Radon Monitors*
S. Oberstedt and H. Vanmarcke, Verhandlungen der DPG Frühjahrstagung Berlin, März 1995

8. *Retrospective Radon Monitoring in Spongy Materials*
S. Oberstedt, H. Vanmarcke, Sixth International Symposium on the Natural Radiation Environment June 5-9, 1995, Montreal (Canada), accepted by program committee

9. *Assessment of radon-daughter deposition in the respiratory tract*
S. Oberstedt, H. Vanmarcke, International Symposium on Radiation Protection in Neighbouring Countries, September 4-7, 1995, Portorož (Slovenia), sent to program committee

10. *Riskassessment via Retrospective Radon Monitoring*
S. Oberstedt, H. Vanmarcke, International Conference on Healthy Buildings in Mild Climate (healthy buildings '95), September 11-14, 1995, Milano (Italy), sent to program committee