

**VOLUME TRAPS -  
A NEW RETROSPECTIVE  
RADON MONITOR**

**S. Oberstedt and H. Vanmarcke**

**Radioprotection  
SCK • CEN**

**BLG-666**

**November 1994**

# VOLUME TRAPS - A NEW RETROSPECTIVE RADON MONITOR

S. Oberstedt and H. Vanmarcke

Studiecentrum voor Kernenergie SCK•CEN, Boeretang 200, B - 2400 Mol, Belgium

## Abstract

A new method to trace back average radon concentrations in dwellings over several decades in time has been developed. This retrospective radon monitor is based on the measurement of the  $\alpha$ - activity of  $^{210}\text{Po}$  deposited in volume traps, e. g. spongy materials used for mattresses and cushions. Polyester samples with different densities have been exposed to radon-laden air. The exposures correspond to characteristic radon concentrations between  $390 \text{ Bq/m}^3$  and  $3.9 \text{ kBq/m}^3$  over a 20 years period. The precision in converting the  $^{210}\text{Po}$ -signal to the radon exposure has been improved by more than one order of magnitude compared to other common techniques. It is shown that this very sensitive method may be applied to almost all types of volume traps used in households.

submitted to Health Physics

## 1. Introduction

In the last decade the radon issue has become one of the major problems of radiation protection. Radon (in fact radon decay products) exposure was recognized to cause lung cancer among mine workers for the first time by Ludewig (1924). However, different surveys showed that high radon concentrations are not only limited to mines and industrial facilities but can also be met in domestic environments due to radon exhalation from the ground (Swedjemark 1987, Green 1992, Poffijn 1992, Kreienbrock 1992). Also in dwellings there is an indication, that an increased lung cancer risk is correlated with high radon exposures. Since it is known that the increased lung cancer risk is due to a radon exposure over a long time period, i. e. several decades, information on the average radon concentration has to be obtained on that large time scale.

In order to monitor the average radon level in dwellings it was proposed to measure the  $\alpha$ - activity of  $^{210}\text{Po}$  implanted in glass surfaces as for example windows and mirrors (Lively 1987, Samuelson 1988). Due to the long half life of  $^{210}\text{Pb}$  (22 y), which decays via  $\beta^-$ -decay through  $^{210}\text{Bi}$  into  $^{210}\text{Po}$ , the long term radon exposure can retrospectively be monitored by measuring the  $\alpha$ - particle emitted from  $^{210}\text{Po}$  with an energy  $E_\alpha = 5.3$  MeV. However, the attachment rate of the airborne radon decay products to aerosol particles and their plate-out rate to indoor surfaces strongly fluctuate depending on the aerosol sources present, e. g. cigarette smoke, cooking. The plate-out is also influenced by the local turbulence and air movement (Vanmarcke 1991). These parameters are retrospectively assessable only with a large uncertainty. Together with the generally unknown cleaning procedures applied to indoor glass surfaces, the precision achievable for this method obtained from existing experimental data (Cornelis 1992, Samuelson 1992) is at best only a factor of about 2.

Therefore, investigations were started at our institute in order to determine the  $^{210}\text{Po}$  activity due to radon decay products deposited in volume traps, e. g. spongy materials used for mattresses and the padding of cushions. Since these materials have a long diffusion length for radon, but are inaccessible for its decay products surface effects are avoided. Thus, the  $^{210}\text{Po}$  signal obtained from the samples is expected to be much better related to the integrated radon exposure.

Some years ago a similar technique was tested on wooden furniture (Falk 1991). However, its

usefulness was limited due to the variable content of natural  $^{226}\text{Ra}$  (and  $^{210}\text{Po}$ , respectively) in the wood. Since more and more spongy synthetic materials are used in houses, which have no such background contributions, this technique became back very attractive.

The aim of our investigations was to determine the calibration factor and to test the reliability of this type of radon monitor. For that purpose radon exposures corresponding to average radon concentrations of typically several hundreds of  $\text{Bq}/\text{m}^3$  over a period of about 20 years had to be simulated in the laboratory. Moreover, the investigated spongy materials should frequently be used in households for mattresses and cushions and should also be of recent fabrication in order to avoid any additional background contribution.

**Table 1:** Characteristic properties of the investigated polyester samples. The error on these values is reported by the producer to be less than 10 % .

no. of sample	density $\rho_s$ ( $\text{kg}/\text{m}^3$ )	rigidity $\kappa_s$ (kPa)
1	20.3	2.8
2	29.3	3.8
3	40.3	4.5
4	44.1	3.1

## 2. Sample selection and experimental setup

For the basic investigations polyester samples ( $\text{C}_6\text{H}_5\text{CH}:\text{CH}_2$ ) with different densities  $\rho_s$  and rigidity  $\kappa_s$  were selected. Their characteristic properties are listed in Tab. 1. The experimental setup is shown in Fig. 1. The radon chamber had a volume  $V_0 = 95.2$  l. Four different samples with a volume of about 6 l each were exposed simultaneously. The air was circulated through a radon source, and the actual radon concentration was continuously recorded with a Lucas-type radon monitor (Lucas 1957). In total 4 runs have been performed with exposures corresponding to average radon concentrations between  $390 \text{ Bq}/\text{m}^3$  and  $3.9 \text{ kBq}/\text{m}^3$  over a 20 years period. After exposure the samples were kept in a radon free environment at least

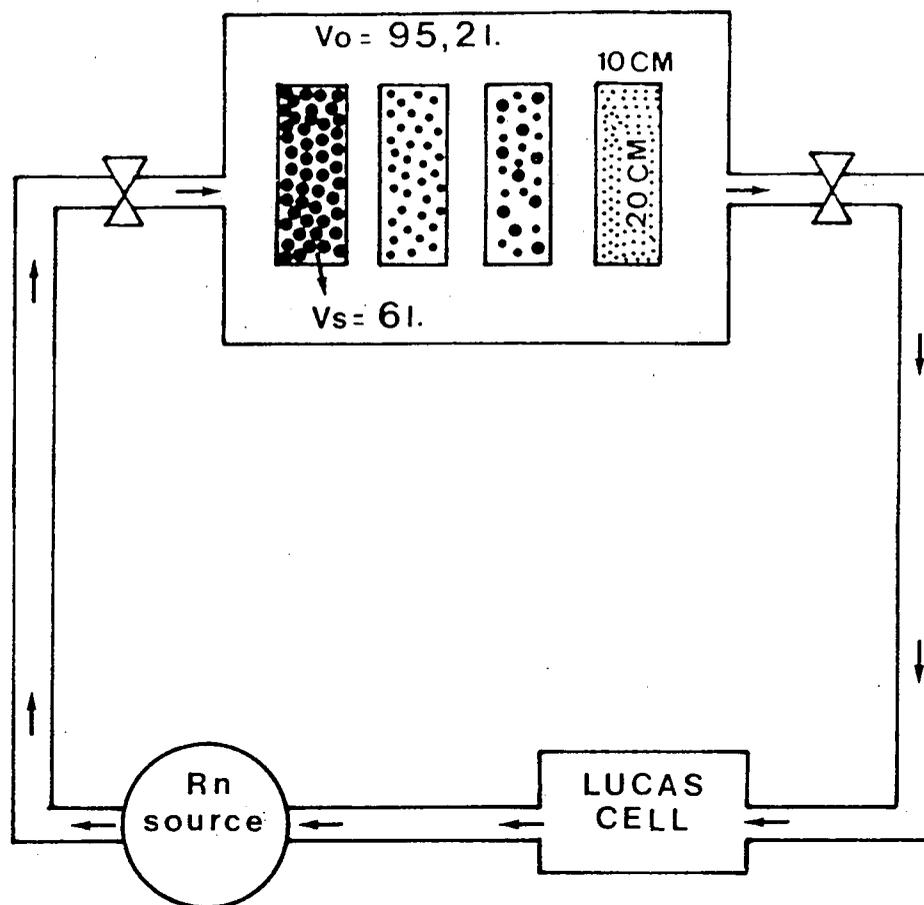


Fig. 1: Experimental setup used for the exposure of spongy material (polyester) to radon.

for about 138 d, which corresponds to one half life of  $^{210}\text{Po}$ , in order to approach radioactive equilibrium. In order to assess the  $\alpha$ - activity, the deposited  $^{210}\text{Po}$  had to be separated from the polyester chemically. For that purpose the exposed samples were cut into smaller blocks with a volume of about  $100\text{ cm}^3$  and an estimated activity of a few tens of mBq. In order to exclude any surface effect these blocks have to be dissected from inside the exposed samples.

### 3. Chemical separation of $^{210}\text{Po}$ and data analysis

The separation of  $^{210}\text{Po}$  from the polyester material consists of different steps. First the small polyester blocks are solved in nitrogen acid (14 molar) and then hydrolized for 5 to 10

hours. After cooling hydrochloric acid (8 molar) is added to the solution and dried under infra-red light. The final residue is solved in hydrochloric acid (12 molar) in order to eliminate all nitrogen acid. The final residue is again solved in 4.2 ml hydrochloric acid (12 molar) and supplemented by water to a final volume of 100 ml. The solution is mixed with ascorbine acid (100 mg) and filled into a plastic bottle, which is covered with a silver plate, turned onto its head and placed near an infra-red light ( $T \approx 60\text{ }^{\circ}\text{C} \dots 80\text{ }^{\circ}\text{C}$ ). During the next 24-48 hours the Polonium autodeposits on the silver plate, which finally is analysed by means of  $\alpha$ - spectrometry.

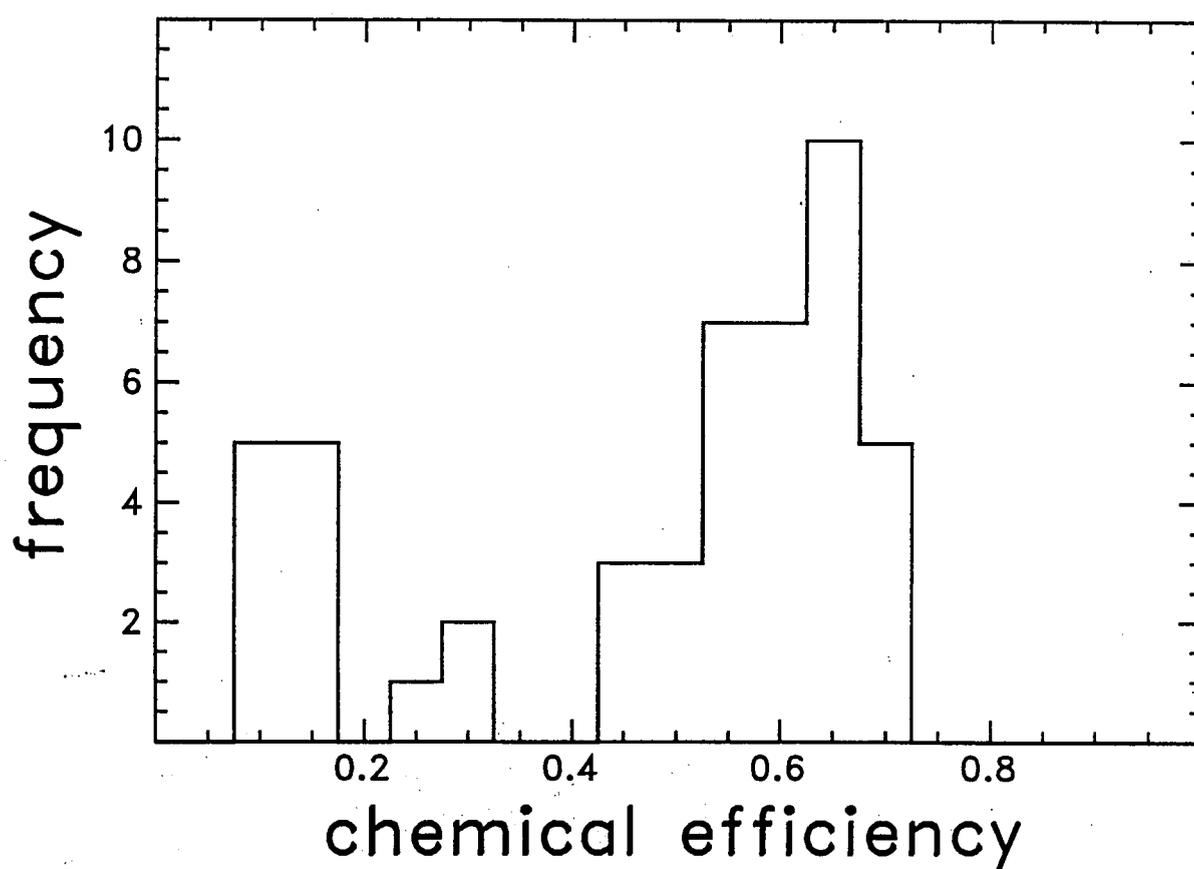
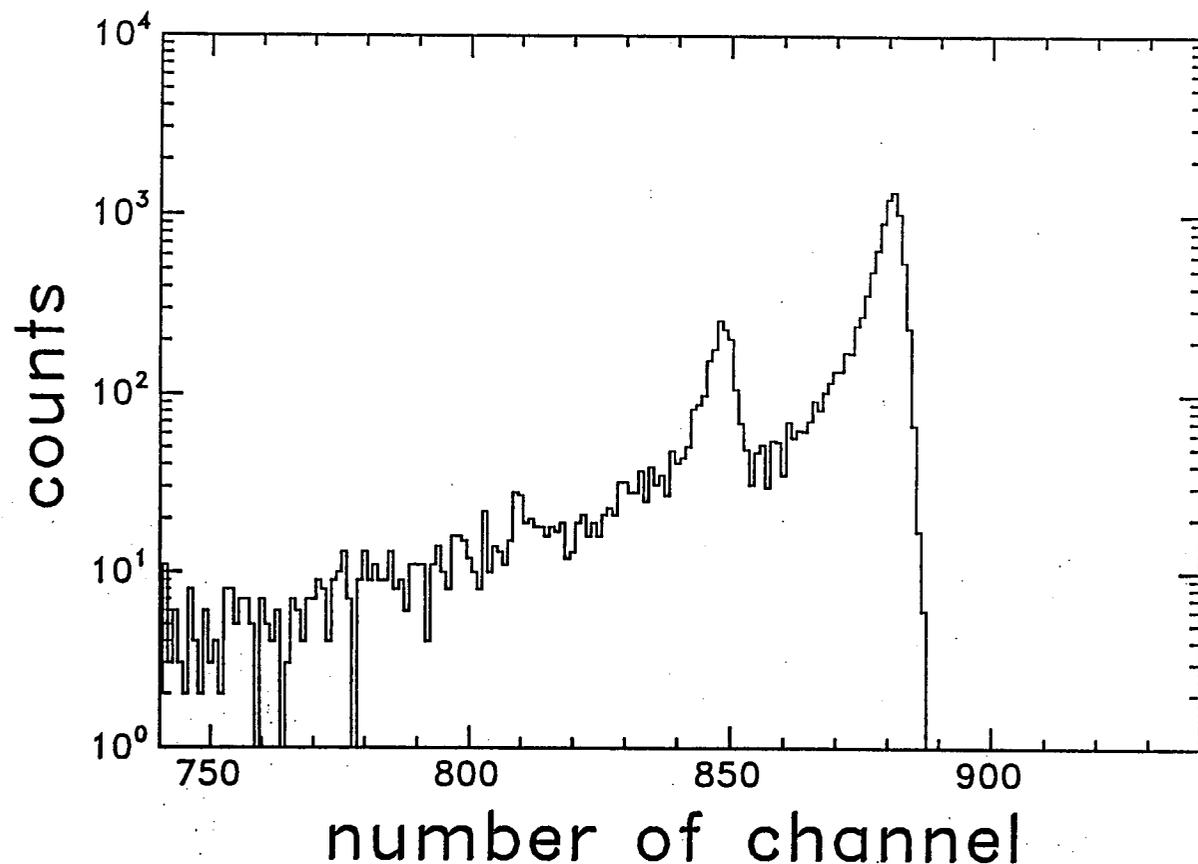


Fig. 2: Frequency distribution of the efficiency of the chemical separation of  $^{210}\text{Po}$ ,  $\epsilon_c$ , deposited in polyester.

The efficiency of this separation procedure is individually monitored by adding a well defined amount of  $^{208}\text{Po}$  as a tracer material. This isotope has a half life for  $\alpha$ - decay of 2.9 y and

emits an  $\alpha$ - particle with an energy  $E_\alpha = 5.1$  MeV. A frequency distribution of the chemical efficiency  $\epsilon_c$  is shown in Fig. 2. The most probable efficiency is about 0.6. The left peak around 0.12 is due to the initial use of less concentrated nitrogen acid.



**Fig. 3:** Typical  $\alpha$ - particle energy spectrum from  $^{208,210}\text{Po}$  deposited onto a silver-plate. The left peak corresponds to the  $\alpha$ - particle at  $E_\alpha = 5.1$  MeV from  $^{208}\text{Po}$  and the right peak to that at  $E_\alpha = 5.3$  MeV from  $^{210}\text{Po}$ .

The  $\alpha$ - activity deposited on the silver plate was then measured in a vacuum chamber using a 900 mm<sup>2</sup> passivated implanted planar silicon (PIPS) detector. The detection efficiency for  $\alpha$ - particles at about 5 MeV was about 17 %. The respective measuring times varied between 40 h for high exposed and 100 h for low exposed samples. A typical  $\alpha$ - particle energy spectrum obtained from these measurements is shown in Fig. 3. The left peak corresponds to

the  $\alpha$ - decay line of the tracer  $^{208}\text{Po}$  at  $E_\alpha = 5.1$  MeV and the right peak to that from  $^{210}\text{Po}$  at  $E_\alpha = 5.3$  MeV.

From this kind of spectra the peak areas were determined in order to obtain the efficiency of the whole procedure ( $^{208}\text{Po}$ ) and the amount of radon progeny ( $^{210}\text{Po}$ ) deposited in the polyester during exposure. The strong tailing on the low energy side of the peaks had to be considered during the peak-area analysis for  $^{208}\text{Po}$ . The analysis of the  $\alpha$ - spectra was carried out with the computer code ALPS 3.11 (Westmeier 1991), which uses a fitting routine according to Slavic (1976) and Westmeier (1990).

Finally, the peak areas for  $^{210}\text{Po}$  were converted to the value corresponding to radioactive equilibrium with  $^{210}\text{Pb}$ .

#### 4. Experimental results and Discussion

For each exposure at least three samples of each polyester material have been processed. The measured  $\alpha$ - activities of each sample were converted into specific sample activities  $a_s$  [mBq/g] at radioactive equilibrium. In Fig. 4  $a_s$  averaged over all analysed samples of each polyester material is given as a function of the radon exposure. Different symbols correspond to different sample densities  $\rho_s$  in the same order as in Tab. 1. Additional measurements on unexposed polyester samples for background determination are included. The error bars extend over one standard deviation. All data sets show a striking linear dependence of  $a_s$  on the radon exposure indicated by the lines in Fig. 4. The linear regression fit parameters are listed in Tab. 2. The uncertainty on the calibration factor, i. e. the slope  $a_1$ , is less than 3.5 %. The main contribution is from the low chemical efficiency achieved during the sample preparation of sample number (1) in Tab. 1. For the other samples the uncertainty is less than 1.6 %.

The calibration factor decreases with increasing sample density  $\rho_s$ . This effect is due to the decreasing ratio of the air volume to the polyester volume. The quality of the data allows to interpolate the calibration factor  $a_1$  for intermediate sample densities with an uncertainty of about the same order of magnitude.

Therefore, the accuracy of this method is higher by more than one order of magnitude compared to techniques relying on the measurement of implanted  $^{210}\text{Po}$  in glass surfaces.

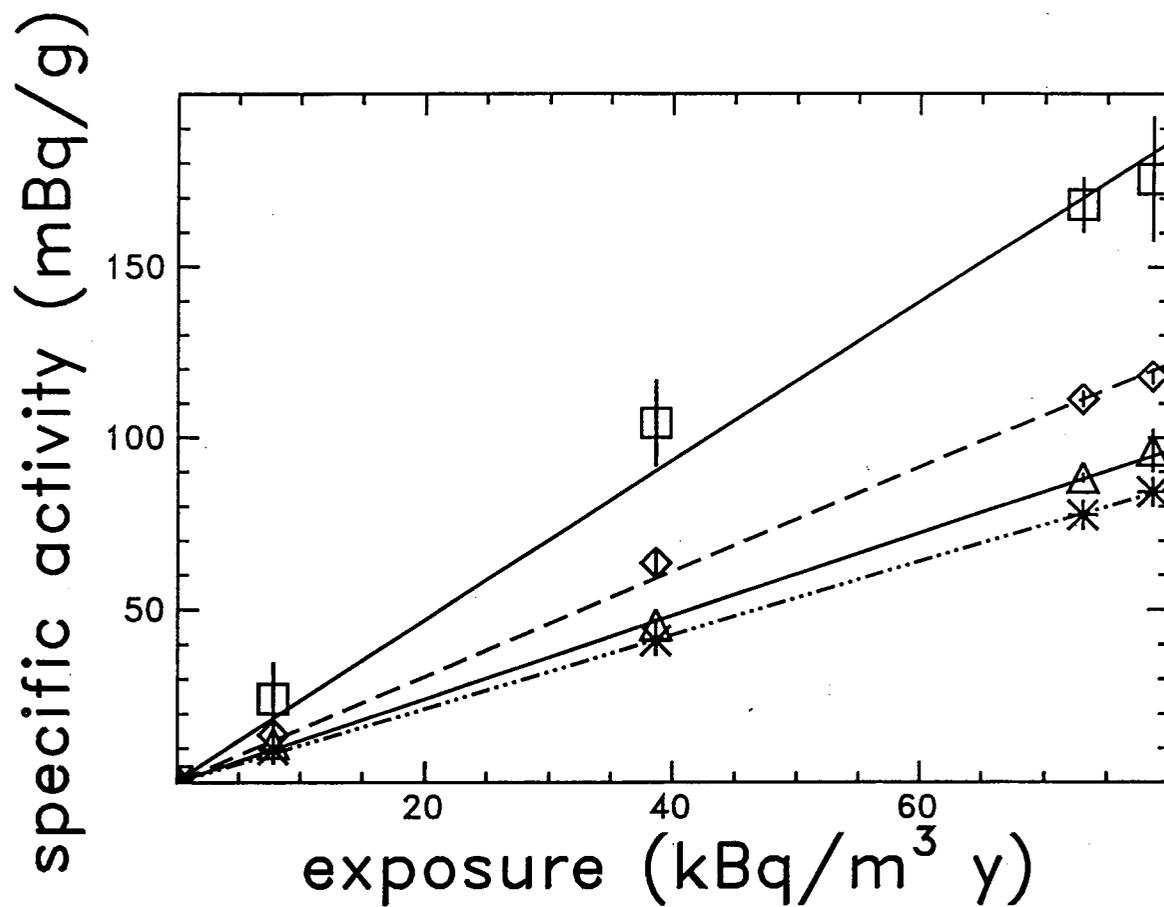


Fig. 4: Specific sample activity  $a_s$  as a function of the radon exposure. Different symbols refer to different sample densities. The order is according to Tab. 2 with (□) corresponding to the lowest density and (\*) to the highest density. The lines indicate the respective linear regression fit to the experimental data.

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

no. of sample	density $\rho_s$ (kg/m <sup>3</sup> )	$a_0$ (mBq/g)	$a_1$ (mBq/g)/(kBq/m <sup>3</sup> 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

As it will be shown next, the experimental data may further be reduced by expressing them in terms of the air volume available in the spongy material. This volume activity  $a_V$  expressed in mBq/cm<sup>3</sup> should be independent of  $\rho_s$ . Furthermore, the conversion factor ( $a_{V(1)}$  in mBq/cm<sup>3</sup>/(kBq/m<sup>3</sup> y)) can directly be calculated.

Assuming an ideal volume trap, i. e. the diffusion length for radon inside is much larger than the dimension of the material, the number of <sup>210</sup>Pb-atoms per 1 kBq/m<sup>3</sup> radon activity concentration,  $N_{Pb}$ , deposited in 1 cm<sup>3</sup> material in a period of one year

$$\begin{aligned}
 N_{Pb} &= 1000 (365.25 \times 24 \times 3600) / (y \text{ m}^3) / \text{kBq/m}^3 & (1) \\
 &= 3.1558 \times 10^4 / \text{cm}^3 / (\text{kBq/m}^3 \text{ y}).
 \end{aligned}$$

At radioactive equilibrium amongst the decay products of <sup>210</sup>Pb the expected conversion factor  $a_{V(1)}^*$  between the <sup>210</sup>Po-activity and the radon exposure in kBq/m<sup>3</sup> y is therefore

$$a_{V(1)}^* = \lambda_{Pb} N_{Pb} \quad (2)$$

$$= 0.0455 \text{ mBq/cm}^3 / (\text{kBq/m}^3 \text{ y}), \quad (3)$$

where  $\lambda_{Pb}$  in eq. (2) denotes the decay constant of  $^{210}Pb$ . Increasing the density decreases the air volume inside the polyester sample so that a correction factor  $k_f$  has to be applied to the experimental data according to

$$k_f = (\rho_m - \rho_s) / (\rho_m - \rho_l) \quad (4)$$

where the density of the sample material is expressed by  $\rho_m$  ( $= 1.05 \text{ g/cm}^3$  for Polyester) and the density of air by  $\rho_l$  ( $= 0.00125 \text{ g/cm}^3$ ). Thus, the conversion factor has to be calculated by

$$a_{V(1)} = \rho_s a_1 / k_f \quad (5)$$

The results for the four samples are 0.0480, 0.0457, 0.0503 and 0.0490  $\text{mBq/cm}^3 / (\text{kBq/m}^3 \text{ y})$ , respectively. In Fig. 5 the derived volume activity is shown as a function of the radon exposure after correction by  $\bar{k}_f = 0.969$ , which is the average correction factor calculated from all sample densities listed in Tab. 2. From the data in Tab. 2 the conversion factor  $a_{V(1)}$  turned out to be

$$a_{V(1)} = 0.0481 \pm 0.009 \text{ mBq/cm}^3 / (\text{kBq/m}^3 \text{ y}). \quad (6)$$

which is in pretty good agreement with the expected value from eq. (3).

This result clearly demonstrates, that this retrospective monitoring technique may be applied to almost all volume traps with a very high accuracy. Together with a systematic error from the determination of the radon exposure the relative uncertainty on  $a_{V(1)}$  is estimated to be smaller than 6 %.

## 5. Conclusion

Our investigations have shown, that volume traps can be used as very accurate as well as sensitive retrospective radon monitors. Under living conditions only a few grams of polyester material have to be acquired in order to get a reasonable information on the long-term radon exposure. In case of polyester materials the detection limit for this monitoring method is 2  $\text{kBq/m}^3 \text{ y}$  at 30 % uncertainty and a measuring period of about 108 h. This corresponds to an average radon concentration of only 100  $\text{Bq/m}^3$  over 20 years. Thus, for epidemiological

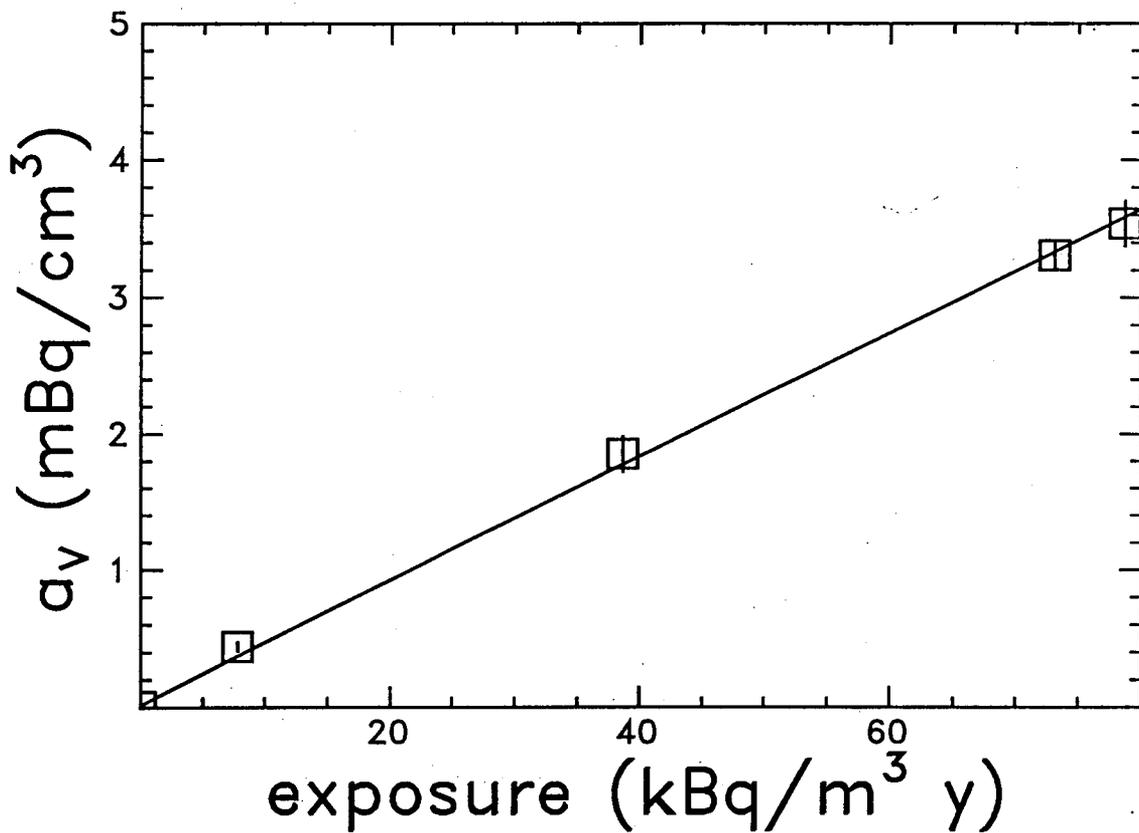


Fig. 5: Volume activity  $a_v$  as a function of the radon exposure. The full line gives the result of a linear regression fit to the data.

studies this method offers a powerful tool to precisely measure the radon exposure retrospectively in dwellings.

During the next phase samples will be selected from houses with a high radon concentration in order to test the method under field conditions and to prove its validity for different types of spongy materials.

**Acknowledgements** - This work was supported by the Commission of the European Communities.

## References

Cornelis J., C. Landsheere, A. van Trier, H. Vanmarcke and A. Poffijn, *Experiments on glass-absorbed Polonium-210*, Appl. Radiat. Isot. Vol. 43, No. 1/2: 127-138; 1992

Falk R., private communication; 1991

Green B. M. R., J. S. Hughes, P. R. Lomas and A. Janssens, *Natural Radiation Atlas of Europe*, Rad. Prot. Dosim. Vol. 45: 491-493; 1992

Lively R. S. and E. P. Ney, *Surface radioactivity resulting from the deposition of  $^{222}\text{Rn}$  daughter products*, Health Phys. 52: 411-415; 1987

Lucas H. F., *Improved low-level alpha-scintillation counter for radon*, Rev. Sci. Inst. 28: 680-683; 1957

Ludewig P. and S. Lorenzer, *Untersuchungen der Grubenluft in den Schneeberger Gruben auf den Gehalt an Radiumemanation* Z. Physik 22: 178-185; 1924

Poffijn A., J. Uyttenhove, B. Drouget and F. Tondeur, *The radon problem in schools and public buildings in Belgium* Rad. Prot. Dosim. Vol. 45: 499-501; 1992

Kreienbrock L., H. E. Wichmann, M. Gerken, J. Heinrich, H.-J. Goetze, M. Kreuzer and G. Keller, *The german radon project - feasibility of methods and first results*, Rad. Prot. Dosim. Vol. 45: 643-649; 1992

Samuellsen C., *Retrospective determination of radon in houses*, Nature 334: 338-340; 1988

Samuellsen C., L. Johannson and M. Wolff,  $^{210}\text{Po}$  as a tracer for radon in dwellings, Rad. Prot. Dosim. Vol. 45: 73-75; 1992

Slavic I. A., Nucl. Instr. Meth. 134: 285ff; 1976

Swedjemark G. A. , A. Burén and Lars Mjönes, *Radon Levels in Swedish Homes : A Comparison of the 1980s with the 1950s*, Radon and Its Decay Products (edited by P. K. Hopke) ACS Symposium Series 331: 84-96; 1987

Vanmarcke H., C. Landsheere, R. van Dingenen and A. Poffijn, *Influence of Turbulence on the Deposition Rate Constant of the Unattached Radon Decay Products*, Aerosol Sci. Technol. 14: 257-265; 1991

Westmeier W. and J. van Aarle, *PC-based high precision nuclear spectrometry*, Nucl. Instr. Meth. A286: 439ff; 1990

Westmeier W, Gesellschaft für Kernspektrometrie mbH, ALPS 3.11 user manual; 1991