COMPARISON OF METHODS TO DETERMINE RADON IN SOIL GAS

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Various methods to determine $^{222}\text{Rn}$ concentration in soil gas were tested at two sites with different soil types in a depth of 1 m. They include instantaneous (spot), continuous (real time) and time-averaging procedures with advective ('active' procedures) or diffusive ('passive' procedures) gas transport from soil to detector: a) active grab-sampling of soil gas from different thin soil gas probes via drying tube and particle filter into evacuated scintillation cells and subsequent analysis of radon activity, b) active sampling by pumping soil gas through continuously operating radon monitors with ionisation chamber or silicon detector and alpha analyser, c) exposition of etch track detectors mounted in small diffusion chambers and, d) diffusion of soil gas into continuously operating, electronic soil probes with silicon detector and alpha analyser.

While the first test site is characterised by its high content of gravel, i.e. a porous soil type, the other test site exhibits a largely impermeable clay soil. The first comparison run was performed during a period of 31 weeks at the site with the porous soil type. Radon concentrations obtained with the different procedures at this site generally agreed within the error limits of the investigated procedures. It has been shown, that installation of probes can influence the observed radon concentration distinct. The second comparison run was performed during a period of 4 weeks at the other site. However, at this site differences of up to a factor of 2 were observed. In addition, sometimes it was not possible at this site to collect a volume of soil gas sufficient for radon analysis. The applicability of the various procedures in the field is discussed.

Keywords: Radon; Soil; Methods; Comparison

INTRODUCTION

Since the main part of indoor radon originates in the soil, knowledge of the radon concentration in soil gas is an important tool to estimate the potential for elevated radon concentrations in new buildings, (e.g. Kemski et al. 1998, Varley and Flowers 1998).

Different methods and systems are available for the collection and measurement of the radon concentration in soil gas. These are instantaneous (spot) measurements, continuous (real time) or time-averaging measurements with diffusive ('passive' procedures) or advective ('active' procedures) gas transport from the soil to the detector (NCRP 1988, Treutler et al. 1998, Hutter and Knutson 1998). The aim of the present investigation was to compare different methods for the determination of radon soil gas with respect to comparability of the results and applicability of the methods in the field. For this, various methods were investigated in two different types of soil: a porous soil with high gravel content and a largely impermeable clay soil. Collection of the soil gas and radon detection was performed in 1 m depth.
MATERIAL AND METHODS

In the present investigation the following methods to determine the radon concentration in soil gas were tested:

- Active grab-sampling of soil gas with a 6 mm external diam. soil gas probe (developed by GSF Research Center for long-term investigations) via drying tube and particle filter into evacuated scintillation (Lucas) cells of 270 ml Volume (model 300A, Pylon) and subsequent analysis of $^{222}\text{Rn}$ activity with a photomultiplier based radon monitor (model AB-5, Pylon). This procedure is selected as the reference procedure. The soil gas probes are permanently installed at the test sites. They were fixed in the soil with sealing foam ensuring that the soil gas being sampled near 1 m depth of the tube end. At the other end, they were equipped with quick connect fittings closed on disconnect.

- Active grab-sampling of soil gas with commercial soil gas probe (model S-2387, 22 mm in ext. diam., Stitz) into evacuated Lucas cells and subsequent radon analysis as described above. These probes are also installed permanently at the test sites.

- Active sampling with the Stitz soil gas probe (model S-2387) by pumping the soil gas through a continuously operating ionization chamber (AlphaGuard PQ2000 pro, Genitron) with an external pump (model AlphaPump, Genitron) at flow-rates of nominal 1 L/min.

- Active sampling with the Stitz soil gas probe (model S-2387) by pumping the soil gas through a continuously operating radon monitor with silicon detector and electrostatic deposition (RTM 2010, Sarad) with internal pump at flow-rates of nominal 3 L/min.

- Active grab-sampling with a commercial 20 mm external diameter soil gas probe (Gammadata) by pumping a total volume of 3 L soil gas through a radon monitor with silicon detector and electrostatic deposition (Markus 10, Gammadata).

- Diffusion of soil gas into continuously measuring electronic soil probes with silicon detector and alpha analyser (Barasol, Algabe and IRG, Tracerlab) Both probes were installed air-tight within 100 mm diameter PVC tubes, for details see Figure 1.

- Diffusion of soil gas into diffusion chambers with etch track detector (Haider et al. 1998) and exposition for a period of 2-3 weeks in the soil. The chambers were installed together with the electronic soil probes as shown in Figure 1.

The distances between the positions where the probes were installed were 1 m each. This distance has been shown to exhibit spatial variations that are low compared with the uncertainty of the methods tested here (Winkler et al. 1999).

The comparisons were performed in two different types of soil:

- During a first period of 31 weeks the radon concentration was measured at a test site with a porous soil with high gravel content located near the GSF Research Centre at Munich-Neuherberg (Germany, Münchener Schotterebene). A more detailed description of this site is given by Bunzl et al. (1998). The $^{226}\text{Ra}$ content is ca. 100 Bq kg$^{-1}$ from 0 to 0.4 m and ca. 30 Bq kg$^{-1}$ from 0.4 to 2.0 m. The $^{226}\text{Ra}$ depth profile is presented elsewhere (Winkler et al. 1999). The top soil depth is about 10-30 cm.

- During a second period of 4 weeks the comparison was performed at a site with a largely impermeable clay soil. It is a building plot situated at Pentling near Regensburg, Germany, and was formerly used as a meadow The top soil is 50 cm deep (Schürzinger 1998). The $^{226}\text{Ra}$
content is ca. 40 Bq kg\(^{-1}\) down to a depth of ca. 1 m and 20 Bq kg\(^{-1}\) from ca. 1.5 m down to 2 m. The \(^{226}\text{Ra}\) depth profile determined at this site is given by Winkler et al. (1999).

RESULTS AND DISCUSSION

The results of the two comparison runs are presented in Tables 1 and 2. The schedules and the various soil radon time series obtained at both test sites are visualised in Figs. 2 through 5.

Comparability

As can be seen in Table 1 and Figure 2, in the porous soil the radon concentrations obtained with the different procedures essentially agree within the error limits of the methods which were estimated to be from about 10% to 25%. In Fig 2, however, it is shown that the radon concentrations obtained with the IRG probe deviate significantly from the results of all other procedures during the period from installation (May 12) to June 10. In addition, this time series exhibits distinct diurnal variations and obviously, the radon concentrations measured at the adjacent positions decreased as a consequence of the installation of the IRG probe. The suspicion of a leak developed during installation of the wide PVC tube needed for the IRG probe (see Figure 1) was confirmed after additional sealing of this tube against the soil at June 10. The electronic Barasol probe (Algade) showed two unexplainable peaks and broke down since the begin of June (Figure 2). Probably due to an incorrect factory calibration of the test unit, the radon concentrations measured with the radon monitor RTM 2010 (Sarad) were too high (Figure 3). After recalibration, this system may also be suitable in this soil type. A separate comparison run was done with the radon monitor Markus- 10 (Gammadata), not available at the time of the first run. As can be seen in Figure 5, on average with this instrument radon concentrations 47% higher than obtained with the reference procedure were observed. Generally, no significant differences between 'active' and 'passive' procedures were detectable in the porous soil at this test site.

As shown in Table 2 and Figure 4 in the largely impermeable soil at the second test site differences between the radon concentrations obtained with the various procedures are obvious. The monitor Markus 10 could not be included in this comparison run because it was not possible to collect the 3 L soil gas, needed for the radon measurement with this instrument. Because of the long time the radon concentration needs to come to the 'true' value after installation, in this soil type 'passive' methods could result in radon concentrations too low compared with 'active' methods (Figure 4). With real time electronic soil probe this increase is monitored and the respective time-integrated radon concentration can be corrected. This correction is estimated to be only about 25% for the IRG probe in the period Nov 26 - Dec 9. However, such a correction is not possible for a time-averaging method, i.e. the etch track detector (I1 and I2 in Figure 3). Nevertheless, at this test site both 'passive' procedures yield soil radon concentrations significantly lower than obtained with the 'active' reference procedure. Whether these differences are due to the way of sampling, i.e. advective or diffusive gas transport from soil to detector, could not yet be clarified. Further investigations are in preparation at this test site.

As can bee seen in Figure 3 the soil radon concentration exhibit distinct temporal variations. The concentrations obtained by short-term measurements, therefore can essentially depend on the time
of measurement. Detailed investigations of spatial and temporal variations of soil radon concentrations are done by e.g. Hutter (1996) and Winkler et al. (1999).

Applicability

The field applicability of the various methods differs essentially:

In both soil types the thin soil gas probes (up to 22 mm in ext. diam.) are very useful, because installation can be easily performed and the disturbance of the soil is minimal. However, in the impermeable soil the active sampling of soil gas with this kind of soil probes was not always possible since the collected soil gas was not always sufficient for radon analysis or water was collected. The time series of the procedures using this type of probes, therefore are incomplete for the impermeable soil (see Figure 4).

On the other hand, the installation of the wide electronic soil probes (up to 75 mm in diam.) and of the etch track detectors within 100 mm - PVC tubes is very labour-intensive. In addition, the soil structure and the soil gas transport can essentially bee disturbed by the installation of this PVC tubes in the porous soil with high gravel content. As noted above the first period of our measurements showed that one of the 100 mm PVC tubes was not installed tight enough. Therefore, even measurements at probes in 1 m distance were disturbed and the measured values were too low (see Figure 2). After additional sealing of the installation tube against the soil only negligible deviations for the various procedures were observed (see Figure 3).

Nevertheless, if they are installed correctly the electronic soil probes are most suitable to measure soil radon concentrations for detailed investigations in real time. The easiest and cheapest way to perform active spot measurements of $^{222}$Rn soil gas concentrations with good precision is offered by the methods which uses thin soil gas probes and evacuated scintillation cells. Methods for the determination of radon gas concentrations, i.e. an the base of ionisation chambers or silicon detectors with electrostatic deposition, in principle are as suitable as the scintillation cell method. However, they are more expensive, especially if more than one or two measurements per site should be performed.

CONCLUSION

Soil radon concentrations obtained with various sampling procedures and detection methods at test site with a porous soil with high gravel content essentially agree within 10%, i.e. within the error limits of the methods. At this site no significant differences were detectable for 'active' and 'passive' procedures, i.e. between advective and diffusive gas transport from soil to detector.

At a second test site exhibiting a largely impermeable clay soil, deviations up to a factor of 2 between the methods were observed, possible due to the different behavior of the soil with respect to the way of the gas transport. Further investigations are necessary at this site.

ACKNOWLEDGEMENTS

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REFERENCES


Table 1: Results of time-averaging measurements in the porous soil (Munich-Neuherberg) with different methods. Methods I1, I2 are etch track detectors installed together with soil probe IRG, B1, B2 with Barasol. For the real time (IRG, Barasol) and the spot measurements (GSF/Pylon 300A, Stitz/Pylon 300A) average values are calculated for the measuring period of the etch track detectors.

<table>
<thead>
<tr>
<th></th>
<th>GSF/Pylon 300A</th>
<th>Stitz/Pylon 300A</th>
<th>IRG</th>
<th>Barasol</th>
<th>I1</th>
<th>I2</th>
<th>B1</th>
<th>B2</th>
</tr>
</thead>
<tbody>
<tr>
<td>22.04. - 4.05.98</td>
<td>9.7</td>
<td>8.1</td>
<td>9.6</td>
<td>10.1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11.05. - 20.05.98</td>
<td>6.6</td>
<td>4.0</td>
<td>4.8</td>
<td>6.9</td>
<td>2.4</td>
<td>7.1</td>
<td>3.0</td>
<td>9.1</td>
</tr>
<tr>
<td>10.06. - 29.06.98</td>
<td>11.0</td>
<td>10.3</td>
<td>10.3</td>
<td>10.4</td>
<td>9.9</td>
<td></td>
<td>10.1</td>
<td></td>
</tr>
</tbody>
</table>

Table 2: Results of time-averaging measurements in the impermeable soil (Pentling) with different methods. Methods I1, I2 are etch track detectors installed together with soil probe IRG. For the real time (IRG) and the spot measurements (GSF/Pylon 300A, Stitz/Pylon 300A) average values are calculated for the measuring period of the etch track detectors.

<table>
<thead>
<tr>
<th></th>
<th>GSF/Pylon 300A</th>
<th>Stitz/Pylon 300A</th>
<th>IRG</th>
<th>I1</th>
<th>I2</th>
</tr>
</thead>
<tbody>
<tr>
<td>25.11. - 8.12.98</td>
<td>59.4</td>
<td>-</td>
<td>22.9</td>
<td>33.0</td>
<td>28.8</td>
</tr>
<tr>
<td>8.12. - 22.12.98</td>
<td>-</td>
<td>60.5</td>
<td>41.3</td>
<td>41.0</td>
<td>41.9</td>
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</table>
Table 3: Adequacy of methods and measuring systems for the determination of radon in soil gas and ratios of estimated values by the different methods to the reference method as obtained for two soil types. Reference method: Lucas cell measurement with grab sampled soil gas (GSF / Pylon 300A-AB 5).

<table>
<thead>
<tr>
<th>soil probe / measuring system</th>
<th>porous soil Adequacy Ratio</th>
<th>impermeable soil Adequacy Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>GSF probe / Pylon 300A-AB 5</td>
<td>++ 1</td>
<td>+ 1</td>
</tr>
<tr>
<td>Stitz probe / Pylon 300A-AB 5</td>
<td>++ 0.99 1.23</td>
<td></td>
</tr>
<tr>
<td>Stitz probe / AlphaGuard PQ 2000pro</td>
<td>+ 0.90 not tested 1)</td>
<td></td>
</tr>
<tr>
<td>Stitz probe / RTM 2010</td>
<td>-2) 1.72 not tested 1)</td>
<td></td>
</tr>
<tr>
<td>Gammadata probe / Markus 10</td>
<td>+ 1.47 4)</td>
<td></td>
</tr>
<tr>
<td>Barasol</td>
<td>+ 1.10</td>
<td>not tested 4)</td>
</tr>
<tr>
<td>IRG</td>
<td>+ 0.94 0.39</td>
<td>+ 0.52</td>
</tr>
<tr>
<td>Etch track detector (CR-39)</td>
<td>+ 0.92</td>
<td></td>
</tr>
</tbody>
</table>

1) The instruments were available only for a short period. Therefore measurements were made only for the porous soil.
2) The test unit was not calibrated correctly. After recalibration this instrument may be useful for this kind of soil.
3) In this soil the internal pump was not able to deliver soil gas.
4) The instrument was defect, the test on the impermeable soil therefore could not be performed.
Figure 1: Installation of soil probes IRG, Tracerlab and Barasol, Allgade together with 2 diffusion chambers containing etch track detectors within a PVC tube. The PVC tube is fixed in the soil and tightened with sealing foam.
Figure 2: Time series of the radon concentrations determined in the porous soil with different methods in the period from 22nd April 1998 to 10th June 1998. Two unexplainable peaks (*) were observed.
Figure 3: Time series of the radon concentrations determined in the porous soil with different methods in the period from 10th June 1998 to 16th November 1998 after additional sealing of the IRG installation PVC tube against the soil.
Figure 4: Time series of the radon concentrations determined in the largely impermeable soil with different methods in the period from 25th November 1998 to 22nd December 1998. On 8th December the etch track detectors were changed and therefore the IRG was removed and reinstalled immediately.
Figure 5: Results of spot measurements of the radon concentration in the porous soil with the system Markus10 (Gammadata) and the reference procedure GSF - soil gas probe / Lucas Cells Pylon 300 A in the period from 4th March 1999 to 1st April 1999.