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Results of the second Dutch national survey on radon in dwellings

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SUMMARY

In 1995 and 1996, radon concentrations and effective air flows were measured in about 1500 Dutch dwellings built between 1985 and 1993. The goal of this investigation was to describe the trend in the average radon concentration and quantify the contributions of the most important sources of radon.

In the living room radon concentrations varied from 5 to 400 Bq m⁻³, with an average of 30 Bq m⁻³, which is 50% higher than the average measured in 1984 in dwellings built before 1970. The average concentration in the outside air was 5 Bq m⁻³ and in crawl spaces 70 Bq m⁻³. The average concentration found in bedrooms was about 10% less than in living rooms.

Measurements of effective air flows showed the most important source of radon in the living room not to be the soil but the building materials, with an average contribution of 70%. The other 30% comprised outside air and air from the crawl space in equal quantities. Use of the mechanical ventilation system was found to slightly reduce the radon concentration in new dwellings. The radon concentration was also found to be positively correlated with the radon concentration in the crawl space and the total area of concrete in the living room.

The long-term increase in the average radon concentration is due principally to the improvements in insulation since 1970, resulting in a fourfold decrease in the building shell permeability, and in a less degree to an increase in the use of concrete by a factor of four over the last 40 years. Since 1970, the average radon concentration in the total Dutch housing stock has increased by about 4 Bq m⁻³, to 23 Bq m⁻³, which is the second lowest average value in Western Europe.

SAMENVATTING

In de periode 1995–1996 zijn radonconcentraties en luchtstromen gemeten in circa 1500 Nederlandse nieuwbouwwoningen (bouwjaar 1985–1993). Het onderzoek had tot doel de trend in de gemiddelde radonconcentratie te beschrijven en het relatieve belang van de diverse bronnen van radon in de woning te kwantificeren.

In de woonkamers varieerde de concentratie tussen 5 en 400 Bq m⁻³. Gemiddeld bedroeg de radonconcentratie er 30 Bq m⁻³ bij een concentratie in de buitenlucht van ongeveer 5 Bq m⁻³. Dit is circa 50% hoger dan wat in 1984 werd gemeten in woningen die tot circa 1970 werden gebouwd. In de kruipruimten is de gemiddelde radonconcentratie 70 Bq m⁻³. In slaapkamers is gemiddeld 10% minder radon gevonden dan in de woonkamer.

Uit meting van luchtstromen in en tussen woonkamer en kruipruimte kon worden afgeleid dat voor de woonkamer de bouwmaterialen de belangrijkste bron van radon zijn met een gemiddelde bijdrage van 70%. Circa 15% van het radon is afkomstig uit de kruipruimte. De buitenlucht levert een even grote bijdrage. De radonconcentratie in de onderzochte nieuwbouwwoningen bleek te worden beïnvloed door het gebruik van de mechanische ventilatievoorziening en gecorreleerd te zijn met de radonconcentratie in de kruipruimte en met het totale betonoppervlak in de woonkamer.

De lange termijn stijging van de radonconcentratie wordt voornamelijk veroorzaakt door verbeteringen in de woningisolatie sinds de zeventiger jaren, wat resulteerde in een vermindering van de luchtdoorlatendheid van de bouwschil met een factor vier, en in mindere mate door een stijging in het gebruik van betonproducten met een factor vier, over een periode van circa 40 jaar. Sinds 1970 is de gemiddelde radonconcentratie voor het totale woningbestand met circa 4 Bq m⁻³ toegenomen tot 23 Bq m⁻³. Dit is de op een na laagste gemiddelde waarde in West-Europa.

1. Introduction

This report describes the results obtained in the second Dutch national survey on radon¹ in dwellings, which was set up with two aims in mind:

- (1) To determine the average and the distribution of the radon concentration in Dutch dwellings built since concluding the previous survey in 1984 [1] and to describe the long term trend. These data will serve as a reference for evaluating the effectiveness of final regulations on radon in new dwellings;
- (2) To quantify the contribution of the most important sources of radon to the indoor radon level in dwellings built in the course of the last decade. These data will be used for the selection of possible countermeasures.

To properly achieve both objects two separate samples of recently built dwellings were taken from the Dutch housing stock: the sample taken for first object was named *SAMPLE N* ('National survey') and for the second object *SAMPLE R* ('Radium study'). Table 1 presents an overview of quantities measured in each of the two samples. More details on the samples and the set-up of the survey may be found in RIVM report no. 610058005 [2], which also contains a description of the equipment used.

Sample R Sample N Quantity measured Radium content of dried soil (Bq kg⁻¹) D Outdoor radon concentration (Bq m⁻³) M M Radon concentration living-room (Bq m⁻³) D D Radon concentration bedroom (Bg m⁻³) D D D Radon concentration crawl space (Bq m⁻³) Effective air flows (m³ h⁻¹) D D S Soil characteristics (various units)

Table 1: Overview of quantities measured

Because the average outdoor radon concentration was expected to be around 3 Bq m⁻³, it was considered very important to adequately quantify the background signal of the radon dosemeters and the levels recorded during transport and storage. Chapter 2 describes tests carried out to this end and their results. Chapter 3 presents the results of the radon concentration measurements. In the subsequent chapters explanations for observed radon levels are sought. The effects of radium content and other soil characteristics on the radon concentration outdoors and in crawl spaces are discussed in chapter 4. The average relative

D Each dwelling where possible

M Eight per municipality

S For all dwellings studied in Maassluis, for none of those in Schiedam, and for four of the dwellings in each of the other municipalities

¹ radon= ²²²Rn in this report

importance of the various sources of radon for its concentration in the living room is dealt with in chapter 5. Three sources will be distinguished: the crawl space, outside air and 'building materials'. The latter source also comprises the hardly contributing minor sources 'use of natural gas' and 'tap-water', which could not be identified separately with the monitoring technique applied. In chapter 6 the effect of various radon-related parameters is studied. The last chapter summarises the main conclusions and discusses policy-related matters.

Throughout this report, the term 'significant' means that a confidence level of 95% was reached. In other words, the criterion for rejecting a working hypothesis in a formal statistical test is p<0.05.

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2. QUALITY ASSURANCE

This chapter deals for the largest part with the experiments that were carried out to test the radon meters and the procedures followed to obtain the measurements. These experiments were necessary for two reasons:

- 1. The second Dutch national survey on radon in dwellings was carried out using the new radon dosemeters from FzK (Foschungszentrum Karlsruhe), whereas KVI (Kernfysisch Versneller Instituut) used the old Karlsruhe cup for the first national survey [1]. The results of both surveys thus could not be compared unless an intercomparison of both instruments and procedures was made.
- 2. The background level of the radon meters had to be quantified to obtain adequate measurements, especially of the radon concentrations in outdoor air. Measurements with the old Karlsruhe cup, for example, revealed that single foils mounted in this cup acquired a much higher number of background tracks than foils stored in stacks in an evacuated cylinder [3].

2.1 FzK dosemeters: tests

All radon dosemeters used in the survey were stored vacuum-sealed in a plastic bag that according to the supplier was 'gas-tight'. This 'gas-tight plastic' consists of three layers: 30 µm polyethylene (PET), 20 µm polyamide and 30 µm PET. A number of aluminium-coated plastic bags were used to test whether these might be less permeable for radon than the 'gas-tight' bags. Several lots of foils, some of which were built into dosemeter housings, were subject to different treatments. Figure 1 gives a short description of these treatments and an overview of the laboratory results. The average track density (cm⁻²) of the different sets of foils were compared at a 95% confidence level, the results of which are summarised below.

Lots 10, 11 and 12

The average track numbers in lots 10 and 11 are significantly higher than in lot 12, for which the following notation is used: [10] > [12] and [11] > [12].

This observation leads to two different conclusions: the background track count increases either (1) due to storage of the vacuum-sealed foils or (2) as a result of a slow shift in the etching process. The latter explanation is assumed to be correct [4]. The etching was done in batches at FzK, which were numbered chronologically. A linear regression analysis of the background foils yields the following relationship between the batch number and the background track density:

$$\left\langle N_{\rm fb} \right\rangle = a_0 + a_1 \times B_{\rm Etch} \tag{1}$$

where $\langle N_{\rm fb} \rangle$ is the expected foil background track density (cm⁻²), $B_{\rm Etch}$ the etch batch number, $a_0 = 2.79 \pm 0.84$ cm⁻² and $a_1 = 1.55 \pm 0.24$ cm⁻².

Lots 1, 2, 3, 4 and 5

Storage 'indoors', as indicated in Figure 1 took place in a C-type radionuclide laboratory with a ventilation rate of 5–10 h⁻¹. Outdoor storage was at 3 m above ground level. Track numbers in foils built into radon dosemeters, sealed in gas-

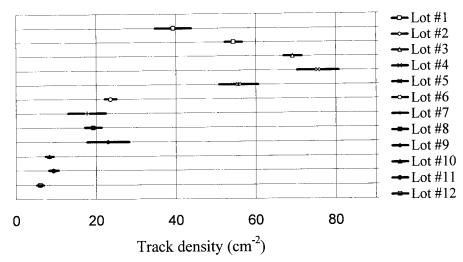
tight plastic and stored indoors (lots 1, 2 and 3) showed a clear increase with storage time ([1]<[2]<[3]). The facts that the increase indoors (from [1] to [3]) compared to the increase outdoors is significantly higher ([3] > [5]) shows that the time-averaged radon concentration indoors is higher than outdoors and that the increase is due to radon leaking through the gas-tight plastic. Sealing the dosemeters with an extra cover of aluminium-coated plastic does not stop the radon leak (not $[4] \neq [3]$). The increase in background tracks on foils built into radon dosemeters, sealed in gas-tight plastic and stored in the C-lab, was described with the following equation derived from lots 1, 2 and 3:

$$\langle N_{\rm S} \rangle = b_0 \times T_{\rm S}$$
 (2)

where $\langle N_S \rangle$ is the background track number (cm⁻²) of dosemeters stored indoors and sealed in plastic, T_S the storage time (d) and $b_0 = 0.140 \pm 0.002$ cm⁻² d⁻¹.

Lots 6 and 7

Comparison of the track numbers in lots 6 and 7 showed no evidence that sealing the dosemeters in gas-tight plastic reduces the track number.



Horizontal lines show 95% confidence intervals, markers show average

Lot #1:16 Dosemeters assembled Feb-Mar 95, stored indoors sealed in "gas tight" plastic, disassembled 31 Oct 95.

Lot #2:187 Dosemeters as in lot #1 but disassembled 31 Jan 96.

Lot #3:185 Dosemeters as in lot #1 but disassembled Apr 96.

Lot #4:187 Dosemeters as in lot #3 but on 31 Jan 96 sealed in an extra cover of aluminum coated plastic.

Lot #5:187 Dosemeters as in lot #3 but stored outdoors from 17 Jun 95.

Lot #6:20 Dosemeters assembled 31 Jan 96, stored indoors sealed in aluminum plastic, disassembled Mar 96.

Lot #7:4 Dosemeters as in lot #6 but not sealed.

Lot #8:4 Dosemeters as in lot #6 but stored in an exsiccator with phospho-gypsum.

Lot #9:4 Dosemeters as in lot #8 but sealed in "gas tight" plastic.

Lot #10:48 Foils, not #assembled, vacuum sealed in "gas tight" plastic stored indoors 31 Jan 96 -- Mar 96.

Lot #11:48 Foils as in lot #10 but vacuum sealed in aluminum coated plastic.

Lot #12:49 Foils as in lot #11 but not stored, immediately sent to FzK instead.

Figure 1: Laboratory results of foils subject to different treatments, with short description of treatments

Lots 8 and 9

Comparison of the track numbers in lots 8 and 9 showed no evidence that sealing the dosemeters in aluminium-coated plastic reduces the track number compared to sealing the dosemeters in gas-tight plastic.

In brief, it can be concluded that the background of the foils depends on the etch batch and amounts to 5.9-12.1 tracks cm⁻². For indoor storage outside the measurement period 0.140 ± 0.002 tracks cm⁻² d⁻¹ must be subtracted. The plastics used for sealing the dosemeters did not significantly reduce the track number.

2.2 Fzk dosemeters: calculation of radon concentrations

The average or expected *net* track density $< N > - < N_{\rm fb} >$ on a foil that was assembled with a radon dosemeter housing from time t_0 to time t_1 and was vacuum sealed for the rest of the time, is proportional to the time-integral of the radon concentration $a_{\rm Rn}$ to which the dosemeter has been exposed, or

$$\langle N \rangle - \langle N_{\rm fb} \rangle = S \times \int_0^1 a_{\rm Rn}(t) dt$$
 (3)

where S is the sensitivity of the dosemeter. If the dosemeter was stored for a period of time, T_S , in a room with an average radon concentration, $\langle a_S \rangle$, was in the back of a car for time T_T (Transport, a_T) and was exposed in a room for time T_E , the average radon concentration during exposure may be derived from equation (4):

$$\langle a_E \rangle = \frac{N - \langle N_{fb} \rangle - \langle N_S \rangle - \langle N_T \rangle}{S \times T_E} \tag{4}$$

where $\langle N_S \rangle = S \times T_S \times \langle a_S \rangle$ and $\langle N_T \rangle = S \times T_T \times \langle a_T \rangle$ are the track densities acquired during storage and transport respectively.

The sensitivity given by FzK is: $S = 0.021 \text{ Bq}^{-1} \text{ m}^3 \text{ d}^{-1}$.

The time periods were obtained as follows:

- 1. The date of assembly was recorded for every radon meter. All radon meters were given to the field personnel for installation on 31 March 1995. The storage time, $T_{\rm S}$, with an average of 33 days, is the number of days between these two dates.
- 2. The transport time, $T_{\rm T}$, with an average of 16 days, is the number of days between 31 March 1995 and the date of placement plus two days for the average transport time after removal of the radon meters.
- 3. The exposure time, $T_{\rm E}$, with an average of 345 days, is the number of days between the date of placement and the removal date.

The time-averaged radon concentration during transport $\langle a_{\rm T} \rangle$ was assumed to be equal to the estimated outdoor radon concentration, being 3 Bq m⁻³. Background, storage, transport and exposure on site were estimated to contribute 3.4%, 2.1%, 0.4% and 94%, respectively, to the average uncorrected radon concentration in the living room.

After having been vacuum-sealed, the foils from the FzK dosemeters were sent to FzK for etching and counting. After that, they were sent back to RIVM together with a list of foil identification numbers and the track densities.

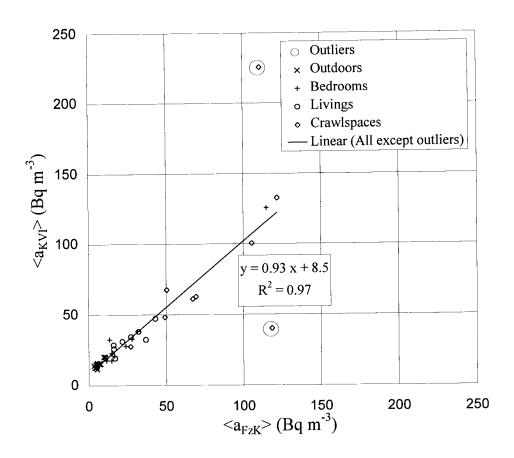


Figure 2: Comparison of KVI and FzK radon meter results

2.3 Comparison of FzK dosemeters and KVI cups

To be able to compare the new survey with the previous one [1], the instruments and procedures applied in both surveys had to be compared. In a number of crawl spaces, living rooms, bedrooms and gardens, both types of radon meters were placed, next to each other. Storage of the FzK dosemeters before they were disassembled was accounted for as described above. The radon cups from KVI were sent to KVI for evaluation. They were received and disassembled up to 20 days later than the corresponding FzK dosemeters. During this time period they were stored outdoors by those who collected the instruments, and assumed to be exposed to a radon concentration equal to the long-term average outdoor concentration, being 3 Bq m⁻³.

KVI, in its covering letter to the track counting results of the Karlsruhe cups, mentioned an extra contribution to the track density. This is proportional to the time that the foils have been inside the cups and corresponds to a radon activity concentration of 7.5 Bq m⁻³ [3]. Furthermore, the authors were aware that the calibration of the KVI cups could also differ from the calibration of FzK dosemeters by a factor f. The radon concentration as reported by KVI, $\langle a_{KVI} \rangle$,

thus may differ from the radon concentration as reported by FzK, $\langle a_{\rm FzK} \rangle$, as follows:

$$\langle a_{\text{KVI}} \rangle = f \times \langle a_{\text{FzK}} \rangle + a'$$
 (5)

with $a' \approx 7.5 \text{ Bq m}^{-3}$.

In Figure 2 values of $\langle a_{\rm KVI} \rangle$ obtained from KVI cups are plotted against values of $\langle a_{\rm FzK} \rangle$ obtained from the corresponding FzK dosemeters. Two points with unexplained large deviations from the regression line were denoted as 'outliers' and excluded from the analysis. Linear regression of the remaining data points yielded:

1. $a' = 8.5 \pm 1.0 \text{ Bq m}^{-3}$, 95% confidence interval (6.4–10.5) Bq m⁻³, and

2. $f = 0.93 \pm 0.03$, 95% confidence interval (0.88–0.99).

The value of a' is not significantly different from the value given by KVI [3]. The value of f, however, is significantly different from 1. As shown by the solid line in Figure 2 both instruments give equal results for radon concentrations around 120 Bq m⁻³.

Figure 2 gives a good impression of the overall random error (σ) in a single radon measurement, which is about 5 Bq m⁻³. The calibration error is probably about 5–10% whereas the error in the factor for comparing the results with those from the KVI cups is about 3%. The two outliers cannot be explained using statistics and are therefore discarded as erroneous results. Both outliers were measurements from crawl spaces. It is possible that, despite all checks, the identification numbers were mixed up.

2.4 Comparison of FzK dosemeters and NRM aerosol monitors

Radon concentrations in outdoor air were measured with FzK detectors at the 11 stations of the Dutch National Radioactivity Monitoring network (NRM) where aerosol monitors are present. As radon concentrations can be derived from data obtained from the aerosol monitors of the NRM [5], a comparison between the FzK dosemeters and the aerosol monitors was regarded as an additional check on the monitoring technique for radon used in this study. For this purpose, all data were converted to EEDC², assuming that the outdoor equilibrium factor is always 0.7 [5, 6] (Figure 3). The two values match poorly for three stations, (numbers 133, 724 and 934), which may be due to the specific local climate leading to a different average equilibrium factor. The FzK dosemeter and the results of the aerosol monitors agree very satisfactorily for the other eight stations, assuming a constant equilibrium factor of 0.7.

 $^{^2}$ EEDC = equilibrium-equivalent decay product concentration, defined as that 222 Rn concentration which, in secular equilibrium with all short-lived decay products represents the same potential α -energy as the mixture actually present.

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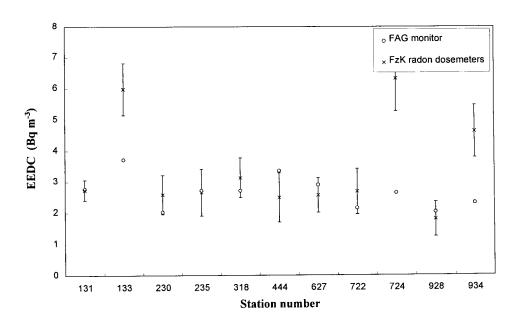


Figure 3: EEDC values in outdoor air at eleven stations of the Dutch radioactivity monitoring network using aerosol monitors and FzK radon dosemeters

2.5 Tracer gas measurements

RIVM registered the link between the sampler code and the address. Linkage of sampler code with analysis number and quality control of the PFT samplers was carried out by Lichtveld, Buis & Partners (LB&P), Utrecht. The samplers were sorted by LB&P and shipped to Brookhaven National Laboratory, New York for analysis. On receipt, the data were checked by LB&P and all erroneous results were discarded.

2.6 Discussion

The quality of the radon measurements with FzK dosemeters was assured by taking the following measures:

- 1. Quantifying the relation between background and batch number, storage time and transport time.
- 2. Assurance of the sensitivity by FzK.
- 3. Comparing results with radon cups from KVI and with NRM data.
- 4. Writing an identification number on the foil as well as on the detector housing. Numbers were registered when placed and when collected. Unrecoverable numbers and duplicate numbers were discarded from the database.

It should be stressed that although a number of statistically sound results may be obtained from this survey given the large numbers of measurements, the value of individual measurements should not be overestimated. The authors recommend to check high values by a second radon measurement before deciding on mitigation activities.

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3. RADON CONCENTRATIONS

This chapter presents the observations for SAMPLE N, a representative sample of dwellings built over the period 1985–1993. Though the data for SAMPLE R, a sample of single-family dwellings built in 1988–1989, are hardly different from those of SAMPLE N, they have neither been included in this presentation nor in the comparison with other surveys and in the analysis of trends (chapter 6). An exception was made for data on bedrooms, which were studied in SAMPLE R only.

3.1 Outdoor radon concentrations

Radon concentrations in outdoor air were measured with FzK detectors in each of the 52 municipalities investigated and at the 11 stations of the NRM equipped with aerosol monitors (see 2.4).

Statistics and a distribution in histogram form of the FzK detector measurements are presented in Table 2 and Figure 4, respectively. The distribution resembles a log-normal form. Geometric mean (GM) and geometric standard deviation (GSD), characteristics of a log-normal distribution, were calculated with the following equations:

$$G M(x) = \exp(\operatorname{average}(\ln(x_i)))$$
 (6)

$$GSD(x) = \exp(\operatorname{stdev}(\ln(x_i))), \tag{7}$$

To test whether the distribution was log-normal the value

$$z_{i} = \frac{\left(\ln(x_{i}) - \ln(GM(x))\right)}{\ln(GSD(x))},$$
 (8)

was calculated for every outside radon concentration measured. If x is distributed log-normally, z should be distributed as N(0, 1), the normal distribution with average zero and standard deviation one. Figure 5 shows the distribution of both z and N(0, 1) for the outside radon concentrations. The data appear to fit in the curve fairly well.

The average value of the measurements near the dwellings is 5.5 Bq m⁻³ (Table 2) and near the monitors, 4.9 Bq m⁻³. Two possible causes for this difference are the higher altitude at which the FzK dosemeters have been placed at the NRM stations (3 m as opposed to 1.5–2.0 m near the dwellings) and their more open situation allowing better mixing of air. The values are low compared to the population-weighted arithmetic

Table 2: Statistics for outdoor radon measurements

| $< a_{Rn}$ | > |
|------------|--------------|
| N | 360 |
| MIN | -0.1 |
| MAX | 23.0 |
| average | 5.5 |
| median | 4.9 |
| N>2 | 357 |
| N>4 | 264 |
| N>6 | 107 |
| N>10 | 15 |
| N>15 | 6 |
| N>20 | 3 |
| GSD | 1.49 |
| GM and 95% | confid. int. |
| GM low | 4.8 |
| GM | 5.1 |
| GM high | 5.3 |

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means of 10 Bq m⁻³ world wide [7], which is explained by the prevalence of sea winds in the Netherlands.

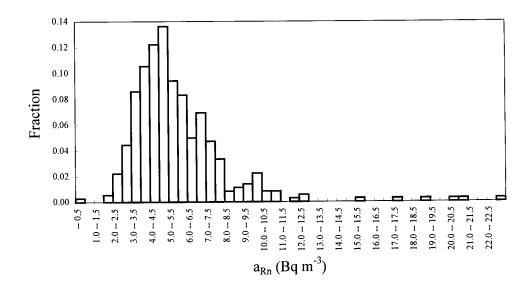


Figure 4: Distribution of outdoor radon concentrations

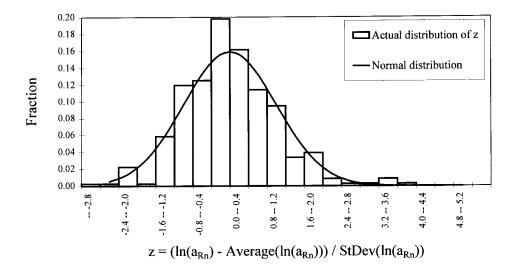


Figure 5: Test for the log-normality of the distribution of outdoor radon measurements

To accurately compare results of various indoor surveys, it is necessary to check and correct for differences in ambient radon level. The data on radon daughter concentrations collected over a period of six years by the NRM [6] were used to ground corrections. The average over the whole period of six years appeared to be significantly lower than the value of the NRM over the study period of the second national radon survey. The average ambient radon level at a height of 1.5–2.0 m above the ground, derived from the six-year period was about

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3 Bq m⁻³, as opposed to about 5 Bq m⁻³ for the period of the radon survey. The long-term average derived from the network is the same as that obtained with the old 'Karlsruhe cups' equipped with two foils and used in the radon survey executed in 1984 [1]. As a result, it was decided to adjust the data of the second survey by subtracting 2 Bq m⁻³ (see Table 5).

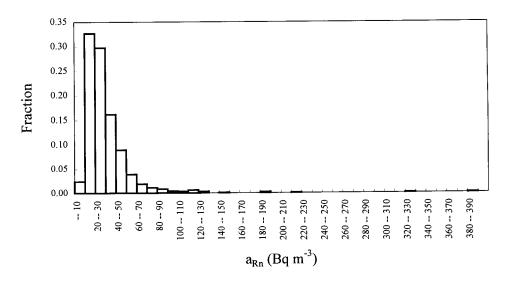


Figure 6: Distribution of radon concentrations in living rooms of SAMPLE N

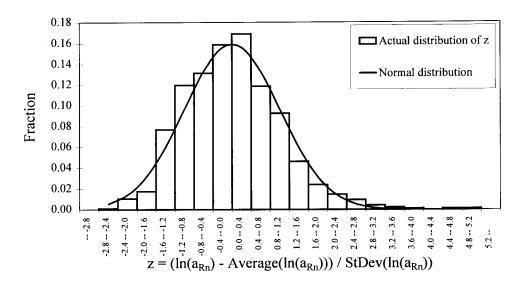


Figure 7: Test for the log-normality of the radon concentration distribution in living rooms of SAMPLE N

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3.2 Radon concentrations in living rooms

SAMPLE N contains 1014 dwellings in which equipment has been placed. Of these, 952 have produced a radon measurement for the living room (Figure 6). In 24 cases, the average exposure time was used because the date of placement or removal of the radon meter was not recorded. The measurements fit a log-normal distribution fairly well, except for a few high values (Figure 7). Especially the two measured above 300 Bq m⁻³ concentrations radon apparently do not fit with the log-normal distribution. Table 3 shows the most important statistics of the radon concentrations in living rooms in SAMPLE N.

The European Commission recommends two action levels for the protection against excessive exposure to radon [8]: (1) for new buildings preventive measures should be taken when levels above 200 Bq m⁻³ are expected, and (2) remedial measures in existing buildings should be taken when levels above 400 Bq m⁻³ are found. Based on the statistics of SAMPLE N, the fraction of new houses with a radon level above 200 Bg m⁻³ is estimated to be 0.012 %, corresponding to about 120 houses built over the past decade. Less than one in a million new Dutch dwellings are estimated to have a radon level above 400 Bq m⁻³. The three measurements above 200 Bq m⁻³ in SAMPLE N, however, suggest that the fraction of dwellings with enhanced radon levels might be somewhat higher. Furthermore, geological anomalies as reported by Albering et al. [9] may cause clusters of enhanced levels, which are either not represented or are underrepresented in the sample studied.

3.3 Radon concentrations in crawl spaces

Figure 8 shows the distribution of the radon concentrations in crawl spaces in *SAMPLE N*. Statistics are given in Table 4, while Figure 9 shows that the distribution resembles the lognormal distribution. The average value of 72.5 Bq m⁻³ is far below the presupposed value of 300 Bq m⁻³ upon which possible mitigation strategies were based in the past [10, 11].

Table 3: Statistics for radon concentrations in living rooms of SAMPLE N

| <a_rr< th=""><th>></th></a_rr<> | > |
|------------------------------------|--------------|
| N | 952 |
| MIN | 5.7 |
| MAX | 381.6 |
| average | 30.3 |
| median | 24.6 |
| N>30 | 335 |
| N>50 | 96 |
| N>100 | 18 |
| N>200 | 3 |
| N>300 | 2 |
| N>400 | 0 |
| GSD | 1.76 |
| GM and 95% | confid. int. |
| GM low | 24.3 |
| GM | 25.2 |
| GM high | 26.1 |

Table 4: Statistics for radon concentrations in crawl spaces of SAMPLE N

| <a<sub>Rr</a<sub> | > |
|-------------------|--------------|
| N | 539 |
| MIN | 8.6 |
| MAX | 510.4 |
| average | 72.5 |
| median | 51.9 |
| N>30 | 409 |
| N>50 | 279 |
| N>100 | 104 |
| N>200 | 27 |
| N>300 | 10 |
| N>400 | 4 |
| GSD | 2.12 |
| <i>GM</i> and 95% | confid. int. |
| GM low | 50.2 |
| GM | 53.5 |
| GM high | 57.0 |

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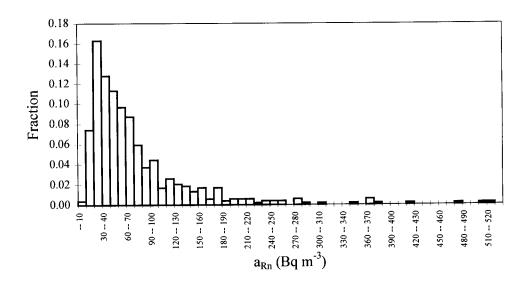


Figure 8: Distribution of radon concentrations in crawl spaces of SAMPLE N

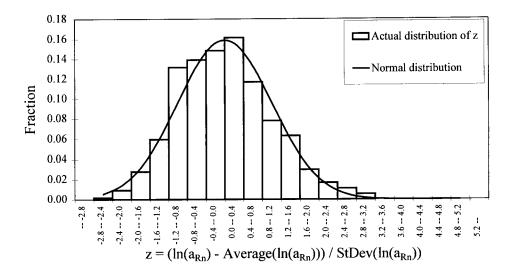


Figure 9: Test for the log-normality of the radon concentration distribution in crawl spaces of SAMPLE N

3.4 Radon concentrations in bedrooms

Concentrations in bedrooms were solely measured in *SAMPLE R*, which is a sample of single-family dwellings and not a random sample from the stock of newly built houses. For the sake of completeness they are, however, given here. Radon measurements were produced for 452 bedrooms. The observed concentrations range from 2 to 265 Bq m⁻³, with an average of 29 Bq m⁻³, which is about 10% lower than the average for the living rooms of *SAMPLE R*.

3.5 Sample-stock corrections

The representativity of the sample for the housing stock was discussed by Hiemstra et al. [2]. The most apparent deviations between stock and sample were the

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over-representation of dwellings built in the second half of the study period, and of private properties and single-family dwellings. Correction for these deviations, however, resulted in negligible and thus unimplemented changes in the data. Other, often unquantified deviations, such as the insufficient representation of old people's flats, were assumed to have minor effects as well.

3.6 Discussion

Because of a systematic error in the measurement technique used, e.g. for the survey of 1984, all Dutch measurements of the indoor radon concentration which have been made with the Karlsruhe cup, are overestimated (see 2.3). To compare the survey of 1984 with the one reported on here, the old data had to be corrected for this error. The monitoring period of the second survey was characterised by a higher than average concentration in outdoor radon. The new data had to be corrected for this deviation before analysis and comparison. There was no need to correct the data of the second survey for deviations of the sample from the original stock.

The consequences of these corrections for both surveys and for each of the locations where measurements were made, are summarised in Table 5. A further analysis of these data is presented in chapter 6.

Table 5: Uncorrected and corrected (geometric) means of in- and outdoor radon levels for the two Dutch national surveys

| Survey and correction | | Radon concentration (Bq m ⁻³) | | | |
|---------------------------|-----|---|--------------|--------|-------------|
| | | Outdoor | Crawl | Living | Bedroom |
| | | air | space | room | (*) |
| First Natl. Survey, | GM | _ | | 24 | _ |
| original data | AM | 3 | _ | 29 | 23 |
| | GSD | _ | _ | 1.6 | - |
| First Natl. Survey, | GM | _ | _ | 17 | _ |
| corrected for | AM | 3 | _ | 22 | 16 |
| measurement error | GSD | _ | - | 1.6 | _ |
| Second Natl. Survey, | GM | _ | 54 | 25 | 24 |
| original data | AM | 5 | 73 | 30 | 29 |
| | GSD | _ | 2.1 | 1.8 | _ |
| Second Natl. Survey, | GM | , | 52 | 23 | 22 |
| corrected for higher than | AM | 3 | 71 | 28 | 27 |
| average conc. outdoors | GSD | _ | - | - | |

^(*) In the second national radon survey, concentrations in bedrooms were obtained from $SAMPLE\ R$ and other concentrations from $SAMPLE\ N$. — means not available

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4. RADON FROM THE SOIL

The radium content of soil samples collected near about 400 single-family dwellings in 14 municipalities and near the aerosol monitors of the NRM³ was analysed [2]. A number of grain-size fractions were determined as well, for about 100 of these samples. These analyses were made to: (1) study the relationship between the radium content in the soil and the radon concentration outdoors and in the dwellings and (2) better predict natural radioactivity levels from soil characteristics.

4.1 Soil radium content and other soil characteristics

Minimum, maximum and average (standard deviation) of the radium content of 475 soil samples from *SAMPLE R* were 6, 72 and 25 (15) Bq kg⁻¹. Most of the locations studied appeared to be built on (supplied) sandy soils hardly varying in radium content. The few locations built on more silty or clayish soils displayed a more variable radium content (Table 6).

Table 6: Average and range of the radium content measured in each study location and averages for the grain size fractions

| Municipality | Mineral 1 | fractions (F) | (mass %) | ²²⁶ Ra (Bq kg ⁻¹) | | | | |
|---------------|-----------|---------------|----------|--|------|------|-------|--------|
| • • | < 2 μm | 250 μm | >50 μm | Average | Min | Max | StDev | Model* |
| Almere | 3.7 | 8.4 | 84.5 | 14.3 | 8.6 | 20.9 | 2.7 | 13.3 |
| Apeldoorn | 3.5 | 14.2 | 79.0 | 15.2 | 10.0 | 20.4 | 2.6 | 14.8 |
| Duiven | 34.7 | 26.4 | 34.6 | 45.0 | 22.1 | 61.0 | 8.7 | 45.2 |
| Gendringen | 12.5 | 12.1 | 72.2 | 26.3 | 15.3 | 38.0 | 6.7 | 22.0 |
| Wisch | 5.3 | 12.0 | 79.1 | 17.2 | 11.7 | 20.2 | 2.2 | 15.7 |
| Nieuwegein | 26.2 | 47.1 | 23.0 | 53.0 | 30.0 | 72.0 | 9.8 | 43.7 |
| Egmond | 3.4 | 8.6 | 85.2 | 10.2 | 5.8 | 14.3 | 2.4 | 13.1 |
| Edam-Volendam | 3.2 | 11.0 | 80.9 | 15.0 | 12.3 | 19.9 | 2.0 | 13.4 |
| Landsmeer | 2.7 | 7.5 | 85.2 | 12.4 | 9.8 | 18.1 | 1.7 | 12.1 |
| Oostzaan | 2.2 | 12.8 | 80.0 | 13.2 | 11.0 | 17.6 | 1.9 | 13.1 |
| Maassluis | | | | 33.4 | 14.8 | 52.4 | 9.6 | |
| Schiedam | 3.5 | 8.4 | 85.2 | 14.8 | 11.3 | 24.4 | 2.3 | 13.2 |
| Asten | 2.7 | 8.0 | 86.9 | 15.1 | 11.4 | 19.5 | 1.9 | 12.4 |
| Voerendaal | 15.4 | 51.6 | 29.6 | 39.5 | 19.4 | 56.0 | 9.2 | 35.6 |

^{*}Value calculated with equation (10).

Köster *et al.* described a linear regression model predicting the natural radio-activity in Dutch soils based on routine soil survey data, in this case the weight percentages of lutum ($< 2\mu m$), silt (2–50 μm) and sand ($> 50 \mu m$) [12]. The equation used until now to predict the natural ²²⁶Ra content is:

²²⁶ Ra_{nat} =
$$(0.60 \times F_{-2} + 0.37 \times F_{2-50} + 0.08 \times F_{50-})$$
 Bq kg⁻¹ (9)

In each of the four sectors of a circular area with a radius of 500 m around a monitoring station of the NRM, 20 evenly distributed samples were taken from the upper 0.5 m of soil. The 20 drillings were combined into one sample in the field.

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This equation proved to be very useful, for instance, for the identification of artificial increases of the radium content [13, 14]. However, since the prediction equation always slightly underestimates the natural radium content (Figure 10), it was necessary to modify its parametrisation on the basis of additional data. The soil data collected for the interpretation of observed radon levels were used to this end. The resulting equation is:

²²⁶ Ra_{nat} =
$$(0.95 \times F_{-2} + 0.36 \times F_{2-50} + 0.08 \times F_{50-})$$
 Bq kg⁻¹ (10)

The authors advise basing future predictions of the level of ²²⁶Ra in Dutch soils on this new equation.

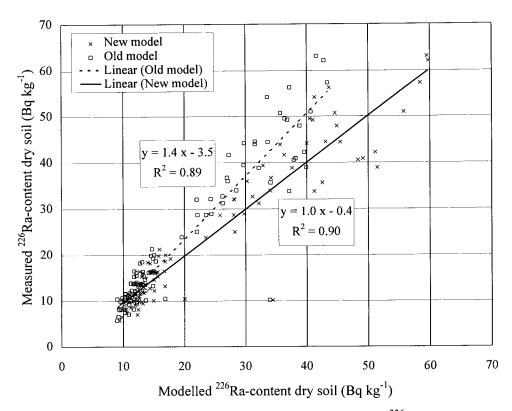


Figure 10: Relation between predicted and measured levels of ²²⁶Ra for the soil samples of the second national radon survey using the prediction equation of Köster et al. [12] and an equation with updated parameter values

4.2 The relation between outdoor radon concentration and soil radium content

The yearly averaged ²²²Rn concentration at the monitoring stations of the NRM clearly relates to the average ²²⁶Ra level in the upper soil layer within a radius of 500 m from the station (Figure 11). This observation confirms earlier ones on the relation between soil radium content of a location and its outdoor radon concentration [1, 15]. It points out the need to use location-specific outdoor concentrations when calculating contributions of the different sources to the indoor concentration (see chapter 5).

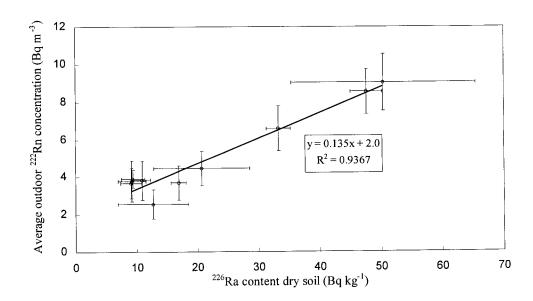


Figure 11: Relation between the average ²²⁶Ra content in the upper 50 cm of soil within a radius of 500 m from a set of FzK detectors and the yearly averaged ²²²Rn concentration derived from these detectors

4.3 The relation between radon in crawl spaces and radium in soil

Linear regression analysis of the 222 Rn concentrations measured in crawl spaces shows a significant positive correlation (although not very strong, $R^2 = 0.39$) with the 226 Ra content in dried soil sampled around the dwellings of *SAMPLE R* (p = 0.02).

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5. VENTILATION AND INDOOR RADON

This chapter presents the results of airflow measurements and discusses the calculations of radon source strengths. Three radon sources will be distinguished: the crawl space, outside air and 'building materials'. The latter source also comprises the hardly contributing minor sources 'use of natural gas' and 'tapwater' [21], which could not be identified separately with the monitoring technique applied. The measurements in multi-family dwellings (N = 91) are not analysed in this context because: 1) the discussion on the effectiveness of countermeasures is strongly linked to the discussion on the relative importance of radon from the soil and 2) most of the newly built houses are single-family dwellings.

However, before dealing with the relative importance of the various radon sources for the concentration in the living room, the grounds for using effective air flows as a characteristic quantity are discussed and airflow measurements for the living room are compared with those reported elsewhere.

| Table 7: Averages and standard deviations of air flows in single-family |
|---|
| dwellings, sample N and Sample R combined |

| Air flow | Number of observations | Average (m ³ h ⁻¹) | Standard deviation (m ³ h ⁻¹) |
|---|------------------------|---|--|
| $\Phi_{\rm c}$ (crawl space total) | 791 | 41.5 | 35.1 |
| Φ_{oc} (outside \rightarrow crawl space) | 728 | 40.9 | 35.6 |
| Φ_{lc} (living room \rightarrow crawl space) | 728 | 1.0 | 3.4 |
| Φ _l (living room total) | 1253 | 85.9 | 45.9 |
| $\Phi_{\rm cl}$ (crawl space \rightarrow living room) | 728 | 6.0 | 6.3 |
| $\Phi_{\rm ol}$ (other \rightarrow living room) | 727 | 78.7 | 43.8 |

5.1 Effective air flow and ventilation rates

Table 7 shows the averages and standard deviations of the air flows obtained from the PFT samplers (see Appendix). The effective air flow from the crawl space to the living room (and vice versa) and the total effective air flow of the crawl space and the living room follow directly from the PFT-measurements. The total effective air flow of the living room is the sum of the flows from the crawl space, from outside and from other parts of the dwelling. Only the first of these three components was measured directly; the partitioning over the last two is unknown. The relative importance of the latter two components will strongly affect estimates of the radon exhalation rate as derived from the radon and ventilation measurements (see 6.3). But it is irrelevant for estimating the relative contribution of the respective sources to the radon level in the living room.

Ventilation is often reported in the form of a ventilation rate, which is the total air flow of a room divided by the volume of the room. Using the volumes of the

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crawl space and the living room reported in the questionnaires, the ventilation rates were calculated. The average result (standard deviation in parentheses) for the living room is $0.9 (0.7) \, h^{-1}$ and for the crawl space $1.1 (1.2) \, h^{-1}$. The variability in the ventilation rate of both spaces is larger than that of the total effective air flow. The range of values for the crawl space is probably wider than that for living room due to inaccurate measurements of the crawl space volume (average of 60 m³).

To assess whether one should see the ventilation rate (total air flow over room volume) as a characteristic quantity or if one should rather report the total air flow as such, a linear regression was done using *SAMPLE N*. The total air flow of the living room was taken as the dependent variable and the volume of the living room as the independent variable. From this test, no dependence was evident, which is consistent with results in a TNO report on an independent set of measurements of the building shell permeability of dwellings [16] (see also 6.4). Therefore there seems to be no ground for using the ventilation rate as a characteristic quantity. As a consequence, neither the ventilation rate nor the volume of any room are included in the equations used in the Appendix.

The TNO data contain measurements of $q_v(10)$, which is the total air flow, Φ , put into a dwelling through a blower door situated at the lowest level and applying a pressure difference of 10 Pa, while all outside doors and windows are kept closed and all inside doors and windows are kept open. Of the 68 dwellings from the TNO data that were built between 1985 and 1995, the average of $q_v(10)$ is 108 L s⁻¹ and the standard deviation 65 L s⁻¹. According to TNO, for a dwelling with a properly working mechanical air exhaust, this value corresponds to an average total ventilation flow during the heating season (wind velocity 5 m s⁻¹, outside temperature 5 °C) of 50 L s⁻¹, or, with the inhabitants present, 70 L s⁻¹, or 250 m³ h⁻¹. The total air flow in the living room is of course smaller than that in the entire building. It is unknown whether the average over a whole year would be larger or smaller than during the heating season. It is therefore difficult to compare the total air flow in the living room measured in our survey with the value of TNO for the total dwelling during the heating season. Nevertheless, with the living-room volume being 30-40% of the total dwelling, a measured average air flow of the living room of 35% of the TNO value for the total dwelling does not seem unrealistic. This observation supports the assumption that the living room is ventilated mainly with outdoor air.

5.2 Radon entry from the crawl space, building materials and outdoors

The dwellings from SAMPLE N and SAMPLE R that yielded a complete set of results were used to quantify the relative importance of the various sources of radon. A complete set of results consists of the radon concentrations for both crawl space and living room, and air flows in and between both compartments. The set contains results of 676 dwellings. The equations used are presented in the Appendix (see also [17]). In these dwellings, on the average, 17% of the radon in the living room comes from the outside air, 14% from sources in the crawl space and the rest from other sources, which are mainly the building materials used for

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the living room (Table 8). Based on the statistical error the crawl space contributes 13.4% to 14.6%, and when based on the total error about 10% to 20%. Similar results have been found in a study by LB&P on measures for reducing the average radon concentration, involving 175 dwellings [18]. The two measures studied by LB&P affect the flux from crawl space to living room and were estimated to decrease the concentration in the living room by about 10% individually or by 15% when applied together.

| Table 8: Radon concentrations and contributions to the radon concentration in |
|---|
| the living room, SAMPLES N and R combined |

| Activity concentration or contribution | Average (Bq m ⁻³) | SEM [*] (Bq m ⁻³) | Contribution to a _L (%) |
|--|----------------------------------|--|------------------------------------|
| a _C : concentration in crawl space | 76 | 2.4 | |
| a _L : total concentration in living room | 32 | 1.0 | 100 |
| a _{1.0} : contribution outside to living room | 5.5 | | 17 |
| a _{L1} : contribution crawl space to living | 4.4 | 0.2 | 14 |
| room a _{L2} : contribution building materials to living room | 22 | 1.0 | 69 |

Supposing that the exhalation rate of the building materials in the crawl space is comparable to that of the materials used in the living room, the relative contribution of radon from the soil to the radon concentration in the crawl space can be derived from data given elsewhere in this report. The data to be used are the average air flow and radon concentration for the crawl space and data on the dimensions of crawl space and living room as registered in the questionnaire (see chapter 6). The average contribution of the soil is estimated to be about $40-55 \text{ Bq m}^{-3}$ or 60-80% of the concentration in the crawl space. This rough estimate provides evidence that on the average building materials cannot be neglected as a source of radon in the crawl space of Dutch houses.

5.3 Regional differences

Regional differences in the relative contributions of the three major sources of radon to the radon concentration in the living room were checked by comparing the averages for the (clusters of) municipalities (Figure 12 and Figure 13). The data of *SAMPLE R* were used to calculate these averages. The basis for clustering certain municipalities is given by Hiemstra *et al.* [2].

If the contributions from the outside air are disregarded, the order of the municipalities hardly changes. Variations in the contribution from the crawl space are relatively large but the absolute contribution is mostly small compared to the contribution from the building materials. The overall picture is clear: roughly 15% of the radon concentration comes from the outside air, 15% from the crawl space and 70% from the building materials above the ground level.

The differences found in the cluster-averaged radon concentrations in living rooms are highly significant, but not straightforward to explain. When plotted on a soil map, only the highest average values of Voerendaal and Nieuwegein seem to coincide with the geology (loam and heavy clay, respectively). The next highest clusters are all in the Western, more urbanised parts of the Netherlands, whereas the lowest three clusters are in more rural areas. Attempts to explain the regional differences using measured air flows and surface areas of concrete fail due to large scatter in the data.

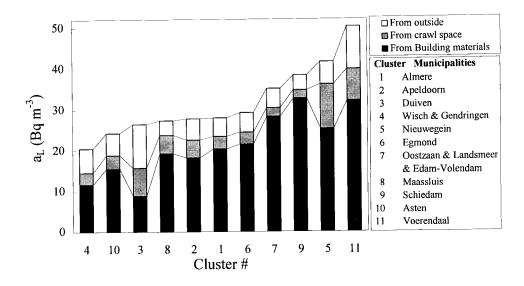


Figure 12: Average contributions of 'crawl space', 'outdoor air' and 'building materials' per cluster of municipalities of SAMPLE R

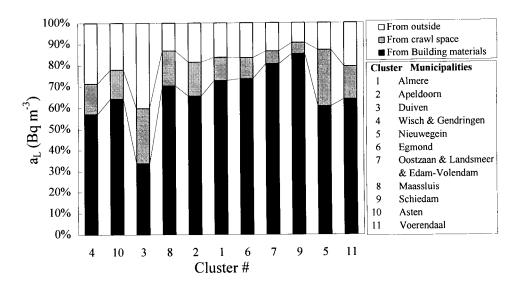


Figure 13: Relative average contributions of 'crawl space', 'outdoor air' and 'building material' per cluster of municipalities of sample R

The significance of the inter-municipality variations in the different contributions should not be overestimated. The average radon concentration in the living room

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is reasonably reliable, since it is the average of many single measurements. The contribution from outside, however, is based on the result of at most eight dosemeters at four locations in a municipality. The contribution from the crawl space is subject to variation because it is derived from the radon concentration in the crawl space, the outside radon concentration and the airflow measurements.

5.4 Environmental quality objectives for the soil

The results of SAMPLE R were obtained to support the further elaboration of environmental quality objectives for the soil with respect to ²²⁶Ra [2, 19]. These quality objectives could then be applied to evaluate artificially enhanced levels of natural radionuclides in soils, such as in polders filled with contaminated harbour sludge in the Rotterdam area. That is why a study of two residential quarters built on harbour sludge was integrated in this survey, i.e. one in Maassluis and one in Schiedam. The size of the samples in each municipality was chosen on the basis of a hypothesised large contribution of radon from the crawl space to that in the living room.

Though higher than expected on the basis of the soil characteristics, the observed ²²⁶Ra levels in the top layer of the soil in Maassluis were not higher than those found in other parts of the country (see Table 6). In Schiedam the radium content of the subsurface layer (-2.0m) was slightly higher than the natural maximum [20], but it was very low in the surface layer (see Table 6). As in other municipalities, building materials were also the most important radon source for dwellings in Schiedam and Maassluis (70–80%) (Figure 12 and Figure 13). As a result, no differences in radon concentration based on differences in ²²⁶Ra content of the soil could be observed for these locations.

It is suggested to use the average radon concentration in the living room, the average radium content of the soil and the average contribution from crawl space to living room as derived from the study as a whole to calculate a conversion factor for assessing contaminated locations. A correction factor for the contribution of radon from the building materials to the radon concentration in the crawl space has to be used (see 5.2).

5.5 Discussion

The contribution of radon originating from the crawl space to the radon level in the living room is much lower than suggested in previous studies [10, 21] (Figure 14). This is mainly explained by:

- 1. A lower than hypothesised radon level in the crawl space.
- 2. A relatively small air flow from crawl space to living room in new dwellings.
- 3. The systematic error in the measuring equipment used in the first survey.

Explanations (1) and (3) are related to each other. In previous studies, the sources 'building materials' and 'outdoor air' were quantified in laboratory experiments and field measurements, respectively. Information on the radon concentration in the crawl space was scarce. As a result, the difference between the radon concentration in the living room as found in the first survey and the contributions

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of the sources 'building materials' and 'outdoor air' was fully attributed to the source 'crawl space'. This difference included the surplus due to the measurement error. Such a line of argument resulted in overestimating both the concentration in the crawl space and its significance for the concentration in the living room.

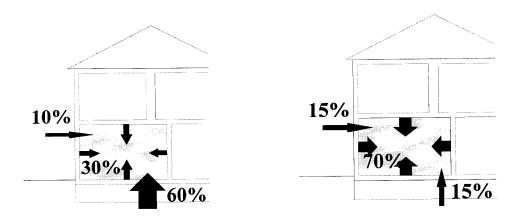


Figure 14: Relative contribution of the major sources of ²²²Rn (building materials, crawl space and outdoor air) to the ²²²Rn level in the average living room determined from previous studies (left), and in this study (right)

The implications of the above results for the effectiveness of countermeasures are:

- 1. For dwellings built in the same way as over the period 1985–1993, further reduction of the influx from the crawl space will reduce the average concentration no more than 15%.
- 2. The most effective measures theoretically are increasing the ventilation of the living quarters with air from outside and using construction elements with a lower radon exhalation rate.

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6. ANALYSIS OF RADON CONCENTRATIONS AND RELATED PARAMETERS

A variety of parameters has been suggested to affect the indoor radon level. Two of the more important ones, soil radium content and ventilation rate, were discussed in the previous chapters. This chapter provides further analysis and interpretation of available results and relates them to those obtained in other studies.

A more detailed description of *SAMPLE N* is first given on the basis of the information from the questionnaire (section 6.1). Next, the potential relevance of this information for the observed radon level is checked (section 6.2). The results are then put into perspective with the results of the first national radon survey [1], a national study on trends in building and demolition waste [22] and an overview of measurements of the permeability of dwellings [16] (section 6.4).

6.1 Description of SAMPLE N using the questionnaire

Measurements were obtained from 1505 dwellings from both *SAMPLE N* and *SAMPLE R*. From 1404 of these dwellings, questionnaires were returned, 962 of which were from *SAMPLE N*. Some of the following data may differ from that given by Hiemstra *et al.* [2], which related to all dwellings of *SAMPLE N*.

The percentage of single-family dwellings in $SAMPLE\ N$ is 87% — the other 13% are multi-family dwellings (apartments). Half of the apartments have their front door inside the building. A further breakdown of the single-family dwellings shows that 23% are detached houses, 22% semi-detached houses, 13% town houses on the corner and 27% enclosed town houses.

Cellars were reported present in 7% of all dwellings of *SAMPLE N*, most of which under part of the building. One-third of the dwellings has no crawl space, and when present it is always under the building as a whole. The crawl space floor is mostly uncovered soil.

Most of the living rooms have an open kitchen and windows on two sides (40%) or an open kitchen and windows on three sides (25%). The living room is in most cases on ground level (88%) and less frequently on the first or second floor (6% each).

The following average living room sizes were obtained (standard deviation in parentheses):

| parentieses). | 2 2 |
|--|-----------------------------------|
| 1. Floor surface area | $39 \text{ m}^2 (12 \text{ m}^2)$ |
| 2. Height | 2.51 m (0.14 m) |
| 3. Net wall area (door and windows excluded) | $77 \text{ m}^2 (20 \text{ m}^2)$ |

The outside walls of the living room were almost always cavity walls (97%), with an outside face of brick masonry (88%). The inner face of these cavity walls was made of sand-lime bricks (47%) or concrete (23%). About the same distribution applies to the materials of the bearing inner walls. The unspecified

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fraction of 30% is divided over the categories 'unidentifiable' or 'other materials'. One or both of the latter categories also accounts for the unspecified fraction of the subsequent characteristics. The non-bearing inner walls are mainly made of gypsum blocks (52%) or gypsum board (11%). The walls are decorated with plaster (21%), wallpaper (27%) or some combination. Most living-room floors are made of concrete (90%), mostly covered with ceramic tiles (24%), wood or cork (24%), or carpet (18%). The floor above the living room is almost always made of concrete (94%). The living-room ceiling is usually plastered.

The inhabitants were asked for the available means of ventilation and their use. Air conditioners and electric fans were almost never used. A considerable number of the extractor hoods are never turned on (34%), most of them only during cooking (60%), while some are always running (5%). Passive exhausts are rare (4%). Windows are used for ventilation about 10% of the time. Small windows are opened more often (25%). The fraction of time that ventilation registers are opened varies from 0 to 100%. About 6% of the dwellings have an open fire place with a chimney which is permanently open. Finally, 45% of the dwellings have a mechanical ventilation system which is always on. Most mechanical ventilation systems are exhaust fans. Only two air supply fans were reported and eight systems for balanced ventilation.

The outside walls of the living room almost always have some form of thermal insulation (97%) and windows have double glass (96%). At least 96% of the dwellings have a central heating system using radiators or floor heating and about 2% have an air heating system. In two cases the air for the air heating system is drawn from the crawl space.

6.2 Relevance of the questionnaire with respect to measured ventilation, radon concentration and radon source strength

Unless indicated explicitly, the observations are based on the data of SAMPLE N. Using linear regression, the numerical answers to questions related to the available ventilation facilities were related to the total flow through the living room as measured with PFT sources and samplers. Examples of such answers are the percentage of time that windows are opened and the area that can be opened. Only two of the items in the questionnaire correlated significantly (p < 5%) with the measured ventilation, which may be rephrased as:

- 1. "Do you have a mechanical ventilation which is always on?" and
- 2. "Do you have a fireplace with a chimney that is always open?".

On the average, a positive response to the first question corresponded to a total airflow which is (21 ± 5) m³ h⁻¹ higher compared to a negative response and for the second question it makes a difference of (27 ± 11) m³ h⁻¹. The average value of the total air flow in the living room is 86 m³ h⁻¹ (see Table 7).

A similar linear regression analysis was done for the radon concentration in the living room. In this case, only the first of the two questions listed above matters significantly. Having a mechanical ventilation which is always on reduces the radon concentration on an average by (3.7 ± 1.7) Bq m⁻³.

A variety of variables from the questionnaire was examined using the statistical techniques *one-way analysis of variance* and *linear regression analysis*. In most cases, log-normally distributed variables such as radon concentrations were transformed to a logarithmic scale before the analysis. The following results were obtained:

- 1. The municipality (11 clusters, Figure 13) matters significantly for the radon concentrations in crawl space, living room, bedroom and outside air (SAMPLE R). No straightforward explanation for the observed differences could, however, be found (see 5.3).
- 2. A significant correlation between the radon concentration in the *crawl space* and in the *living room* was found (p<0.0005), but *not* between the radon concentration in the *crawl space* and in the *bedroom* (p=0.091) (SAMPLE R). The concentrations in the living room and the bedroom were significantly correlated (p<0.0005) (SAMPLE R).
- 3. No significant effect of the presence of a crawl space on the radon concentration in the living room of single-family dwellings was found.
- 4. A significant correlation between the radon concentration in the outside air and the radon concentration in the living room was found (p<0.0005).
- 5. No significant difference was found between the average radon concentration in the living room of single-family dwellings and of multi-family dwellings (apartment buildings), which is different from the situation during the first national survey [1]. This aspect of the study will be elaborated in 6.4.
- 6. No significant effect of the floor level of the bedroom on the radon concentration in the bedroom was found (SAMPLE R).
- 7. There is no convincing relation between the date of construction and the radon concentration in the living room.

6.3 The radon source 'building materials'

The radon source strength in the living area, S_L , was calculated using equation (22) shown in the Appendix. The average value of S_L is 530 mBq s⁻¹. In this section an attempt is made to relate this source strength to the building materials reported in the questionnaire.

From a study by Intron [23], in which radon exhalation rates of six types of building materials were measured, building materials may be roughly divided into four categories according to their average radon exhalation rate, E:

- 1. Concrete: $E_{co} \approx 0.7-2.9 \text{ mBq m}^{-2} \text{ s}^{-1}$
- 2. Sand-lime bricks: $E_{sl} \approx 0.35 \text{ mBq m}^{-2} \text{ s}^{-1}$
- 3. Bricks, cellular concrete and gypsum blocks: $E_{bg} \approx 0.10 \text{ mBq m}^{-2} \text{ s}^{-1}$
- 4. Other materials (wood panels, gypsum board, etc.): $E_{om} < 0.02 \text{ mBq m}^{-2} \text{ s}^{-1}$.

In principle, the radon source strength is the sum taken over these categories of the average radon exhalation multiplied by their surface area in the living area. However, if in the questionnaire a construction element was reported to consist of a combination of building materials or if the material was unknown (about 5% of the total surface area), it was assigned to category (4). Therefore, this category

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contributes an unknown amount to the total radon source strength. Average areas of the four categories are presented in Table 9. When calculated this way, the source strength in the living room ranges from 80 to 300 mBq s⁻¹, which is at least a factor 1.7 below the value of S_L (530 mBq s⁻¹) derived from our measurements. In other words, at least 40% of the derived radon source would be missing.

As was suggested in 5.1 the total air flow of the living room does not consist of outdoor air only, but comes in part from the rest of the dwelling. Carried to the extreme, this would imply that the measured air flow represents the flow of the entire dwelling and that all building materials contribute to the radon concentration of the living room. If this is the case, the amount of building materials contributing would be at most a factor 2.5 higher. It may explain most, if not all, of the source which is missed when only the building materials in the living room are considered.

Table 9: Average surface areas (m^2) of building materials of four categories in living rooms of SAMPLE N

| Area (m ²) | Total area | Concrete | Sand-lime brick | Clay bricks | Others |
|---------------------------|---------------|----------|--------------------|----------------|--------|
| Floor and ceiling | 80 | 80 | 0 | 0 | 0 |
| Outside walls, outer face | 32 | 1 | 2 | 28 | 1 |
| Outside walls, inner face | 32 | 4 | 18 | 3 | 7 |
| Bearing inner walls | 26 | 12 | 9 | 3 | 2 |
| Non-bearing inner walls | 20 | 0 | 0 | 14 | 6 |
| Total | 190 | 97 | 28 | 48 | 16 |

To see if the information on building materials collected with the questionnaire relates to the observed source strength, it is also possible to carry out a linear regression with the derived source strength, S_L , in the living room (see Appendix) as the dependent variable and the surface areas of all categories (except category 4) as the independent variables. The coefficients found represent in principle the average exhalation rates of the categories.

Due to a large amount of scatter ($R^2 = 0.2$), no significant values for categories 2 and 3 could be found, but the value for category 1 (concrete) is significant (p = 0.01), with a value of 3.2 ± 1.2 mBq m⁻² s⁻¹. The intercept of 70 ± 40 mBq s⁻¹ is not significant (p = 0.07) and small compared to the average contribution of concrete ($97 \text{ m}^2 \times 3.2 \text{ mBq m}^{-2} \text{ s}^{-1} = 310 \text{ mBq s}^{-1}$). Both the laboratory measurements of the exhalation rate of concrete and the value derived from the linear regression show large variations. The latter value is, given its uncertainty, within the upper end of the range reported by Intron [23]. Because an unknown fraction of the air flow does not come from outdoors but from the rest of the dwelling, the average exhalation rate will be lower by at most a factor of 2.5.

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6.4 Trend analysis

Figure 15 shows the average radon concentration in living rooms for the total Dutch housing stock and for all dwellings built in a given decade after 1930. In this figure, the corrected results of both the first and the second national surveys (see 3.6) were combined with data from Central Bureau of Statistics on the numbers of single- and multi-family dwellings built in each decade. The radon concentration in houses built since 1980 is roughly 50% higher than in houses built before 1970, which is reflected in an increase of about 20% in the average over the total housing stock. The changes in building practices before and after the seventies had minor effects in terms of indoor radon concentration in comparison with the changes introduced in the seventies. If the building practice of the last decade is continued, the average radon concentration for the total housing stock will gradually increase up to an asymptotic value of 28 Bq m⁻³.

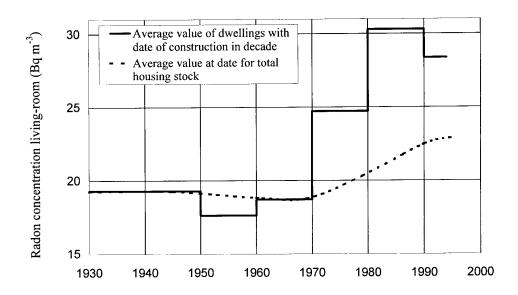


Figure 15: Average radon concentration in living rooms for the total Dutch housing stock and per decade

Figure 16 shows the same data for single- and multi-family dwellings separately. In dwellings built since 1984, radon concentrations in the living rooms of single-and multi-family dwellings are not significantly different. In old dwellings the radon concentrations in the living rooms of multi-family dwellings (N = 181, $a_{Rn} = 25.2 \text{ Bq m}^{-3}$) are consistently lower than in single-family dwellings (N = 455, $a_{Rn} = 29.4 \text{ Bq m}^{-3}$). The 95% confidence interval of the difference is 1.8–6.6 Bq m⁻³. When the soil is an important radon source, this pattern is to be expected, as the influence of this source will decrease with increasing floor number. In multi-family dwellings, the living room is often above and in single-family dwellings mostly at ground level. The soil is therefore likely to be a more important source of radon in old dwellings, as opposed to new dwellings, where, as we have concluded, most of the radon comes from the building materials.

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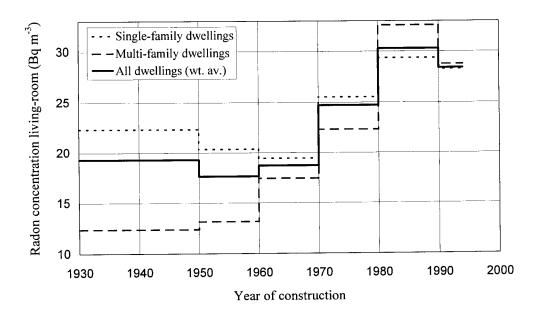


Figure 16: Average radon concentration per decade in living rooms of singleand multi-family dwellings

Two trends can be observed: (1) radon concentrations are increasing, and (2) a larger part of the radon in single-family dwellings would come from the building materials. Observation (2) seems easiest to explain: in the Dutch Building Decree, a minimum air-tightness of the ground floor of new buildings is prescribed. This, in combination with new construction techniques, probably lead to ground floors with a higher air-tightness than before. The increasing radon concentrations (first observation) may be caused by two developments.

The first is an increasing air-tightness of the building shell of new dwellings and a change in building materials used. According to a study by TNO, the air-tightness in Dutch dwellings has increased gradually since 1970–1975: the air-tightness of dwellings built in 1990–1995 is four times higher than of dwellings built before 1970 (Figure 17) [16].

The second possible cause of the increase in the average radon concentration in new dwellings is a change in building materials. It is illustrated in Figure 18 which is derived from data published by De Graaf [22]. This figure shows that the use of concrete steadily increased over a period of about four decades. It progressively substituted wood used for the construction of floors, and bricks used for the construction of walls. The use of sand-lime bricks and gypsum blocks for inner walls caused a further reduction in the use of clay bricks. For multi-family dwellings the changes after 1945 are less pronounced.

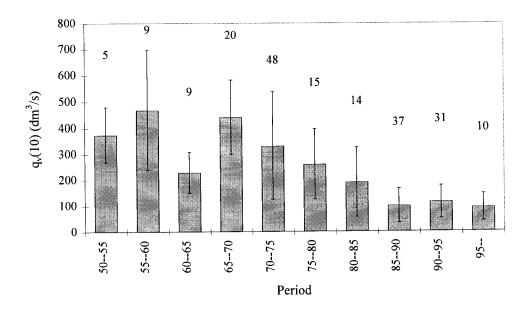


Figure 17: Average and standard deviation of the air tightness of dwellings as a function of year of construction and number of dwellings per period [16]

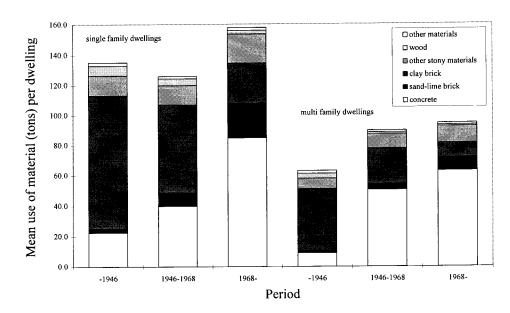


Figure 18: Mean use of six categories of building materials in single- and multifamily dwellings for three periods of time (wood and concrete used for piles has not been included in this figure)[22]

The relative importance of both suggested causes of the observed increase is hard to quantify. The use of concrete, which is the dominant building material in terms of radon exhalation, and the air-tightness both increased by about a factor of four, resulting in a maximum combined effect on the radon concentration of a factor of 16. The observed increase in indoor radon by 50% over the period 1930–1995 is by far smaller than this theoretically maximum effect. This discrepancy suggests that the contribution from crawl space to living room was much larger in old dwellings, as has been argued elsewhere in this section. Furthermore, the

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decrease in the actual ventilation is probably not as large as the decrease in $q_{\nu}(10)$, because older dwellings are more often ventilated passively, while most new dwellings have a mechanical ventilation system. The relevance of the type of building materials for the build-up of a certain radon level in dwellings is apparent from another study of TNO on a limited number of new houses [24]. It shows dwellings with a timber frame to have a lower indoor radon level than conventional types of dwellings. The congruence between the change in airtightness and in indoor radon level in the seventies suggests, however, this factor to be the most important.

6.5 Discussion

In this chapter the influence of various building characteristics on the indoor radon concentration was analysed. This was done by analysing the questionnaire and by combining the data of the two Dutch radon surveys with those of some national studies on building characteristics. Each of the approaches suggests that both ventilation and building materials affect the radon concentration. The relative importance of both causes of the observed increase is, however, hard to quantify.

The data of the questionnaire reveal that ventilation rate and surface area of concrete in the living room relate to the radon concentration. Most manifest in the long-term trend of the indoor radon concentration are the pronounced increase of the radon level in the seventies which levels off in the eighties, and a disappearing difference between single-family and multi-family dwellings. The latter observations supports the hypothesis of a progressive decrease of the influence of the soil as a source of indoor radon. The studies on building characteristics suggest that mainly changes in ventilation characteristics caused the shift in indoor radon level.

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7. CONCLUSIONS AND IMPLICATIONS OF THE SURVEY

7.1 Main conclusions

The main conclusions on the total Dutch housing stock are drawn from observations concerning *SAMPLE N* (dwellings built over the period 1985–1993), put into perspective with the results of the first national survey on dwellings built up to 1984 [1].

- 1. Due to an error in the measuring equipment used in the first survey, the average radon concentration in dwellings has been overestimated by approximately 7 Bq m⁻³. The corrected average radon concentration in living rooms, measured in the first survey is 22 Bq m⁻³.
- 2. During the measuring period outdoor radon concentrations were higher than average. This called for a correction of 2 Bq m⁻³ when comparing the new data with data from the previous survey.
- 3. The average radon concentration in living rooms and crawl spaces of new dwellings (built 1985–1993), corrected for the higher than average outdoor concentration, is lower than expected: 28 Bq m⁻³ and 70 Bq m⁻³, respectively. Expected concentrations were 35 Bq m⁻³ [25] and 300 Bq m⁻³ [10], respectively. The differences between prognosis and measurement are in part explained by the systematic measurement error in the first survey.
- 4. The contributions of outdoor air and of sources in the crawl space are both approximately 15%. The remaining contribution of 70% from building materials is much larger than stated in the Integrated Criteria Document on Radon [21].
- 5. The difference in the average radon concentration in living rooms in single-family dwellings compared to apartments, which was observed in the first survey, is not present in the data of the second survey.
- 6. There is a clear trend in the radon concentrations in both single- and multifamily dwellings. The average in new dwellings is roughly 50% higher than in dwellings built before 1970. The average in the total housing stock has increased about 20% since 1970 up to 23 Bq m⁻³. If the building practice of the last decennium is continued, the average radon concentration for the total housing stock will gradually increase up to an asymptotic value of 28 Bq m⁻³.
- 7. Based on the statistics of *SAMPLE N*, the fraction of new houses with a radon level above 200 Bq m⁻³ is estimated to be 0.012 %, corresponding to about 120 houses built over the past decade.
- 8. The increased air-tightness of the building shell since 1970 and a progressive change in the use of various building materials may explain the observed trend in the indoor radon concentration. Though the relative importance of both suggested causes is hard to quantify, the first factor is thought to be the most important one.

The main conclusions from the "radium-radon" study (SAMPLE R) are:

- 1. Radon concentrations in all compartments (living rooms, bedrooms, crawl spaces and outdoors) vary significantly over the municipalities studied.
- 2. Radon concentrations in living rooms averaged over municipalities differ by

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more than a factor of 2.5. The causes of the observed variation remain to be explained.

- 3. The contribution of building materials to radon concentrations in living rooms averaged over municipalities is 10–30 Bq m⁻³ or 35–85%.
- 4. No significant effect of the presence of a crawl space on the radon concentration in the living room was found.
- 5. Making use of the average radon concentration in the living room, the average radium content of the soil and the average contribution from crawl space to living room as derived from the study as a whole is suggested to calculate a conversion factor for the assessment of contaminated locations. A correction factor for the contribution of radon from the building materials to the radon concentration in the crawl space has to be used.

7.2 Implications for dose estimates

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Following the recommendations of ICRP, an average 222 Rn concentration of 1 Bq m⁻³ in the dwelling corresponds to a potential alpha energy concentration of 1.56×10^{-2} mJ h m⁻³ a⁻¹, assuming an equilibrium factor of 0.4 and an indoor residence of 7000 h a⁻¹ [27]. In combination with a dose conversion coefficient for 222 Rn of 1.1 mSv per mJ h m⁻³, the effective annual dose rate is 17.2 μ Sv a⁻¹ per Bq m⁻³ [26, 27].

The average indoor radon concentration for the total housing stock in 1995 was 23 Bq m⁻³. Based on the risk factors published by ICRP [27], this would induce 29 cases of lung cancer per million per year, which corresponds to about 450 cases per year in the Netherlands. The estimated number of *extra* lung cancers in 1995 due to the increase in radon concentration since 1970 is about 75 in the Netherlands (5 per million) per year.

The new risk estimate for the actual indoor concentration of ²²²Rn for the total housing stock is far lower than the number of 48 cases per million of inhabitants and per year as reported in 1993 by Vaas *et al.* [21]. This is partly due to the fact that Vaas *et al.* based their calculations on the results of the indoor radon survey of 1984, in which the radon level was overestimated by about 30% (see section 2.3), and partly to a gradual changes in the dose conversion coefficient and the risk factor for radon as recommended by ICRP.

7.3 Recommendations for future research

Prescription of any radon-related measure deserves:

- an explanation of the discrepancy between the estimate of the exhalation rate
 of building materials, i.c. concrete, as derived from this survey and from
 measurements made in the laboratory. This discrepancy can be solved by a
 more detailed and careful analysis of the lab measurements, amended with
 new measurements if needed, and by model calculations on ventilation and
 radon transport in dwellings;
- 2. more decisive information on the relative importance of changes in the airtightness of dwellings and in the use of building materials for the indoor radon level. Model calculations linked to results of this survey may

- significantly contribute to answering this question;
- 3. some additional measurements on the relative importance of the radon sources 'soil' and 'building materials' in old dwellings. This information is needed to ground radon-related countermeasures in existing dwellings;
- 4. a survey to check the effectiveness of implemented countermeasures.

7.4 Implications for policy making

The national policy on radon in the Netherlands for new dwellings at present focuses on the introduction of a performance level for the radiation dose. For existing dwellings it is based on measures taken within the framework of sustainable building. The implications of the results of this study are:

- 1. Providing crawl space ventilation and making the ground floor air tight seems still feasible for existing dwellings, where the contribution from the crawl space is probably higher than in new dwellings.
- 2. Focusing more on adequate ventilation of the living quarters for both new dwellings and dwellings to be renovated as an effective measure for controlling and reducing the radon concentration.
- 3. Besides focusing our on attention to ventilation and ground-floor insulation, the choice of building materials will affect the radon concentration.

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APPENDIX: THE THEORY OF RADON TRANSPORT AND SOURCES AND THE USE OF TRACER GAS MEASUREMENTS

Tracer gases

Two types of perfluorcarbon tracer (PFT) gas were used: perfluormethylcyclohexane (PMCH) and perfluordimethylcyclohexane (PDCH). In most dwellings, PDCH was released in the crawl space and PMCH in the living room. There were, however, a few exceptions.

In the equations the gas released in the crawl space is labelled 'C', and the gas released in the living room 'L'. In the following list of symbols, the first six are measurement results, the next four are calculated directly from the measurement results and the last two are derived from the calculated quantities.

Measured $P_{\rm C}$ Tracer gas ('C' type) produced hourly in the crawl space

P_L Tracer gas ('L' type) produced hourly in the living room

 $C_{\rm CC}$ Concentration of 'C' in crawl space

 $C_{\rm CL}$ Concentration of 'C' in living room

 $C_{\rm LC}$ Concentration of 'L' in crawl space

 $C_{\rm LL}$ Concentration of 'L' in living room

Calculated Φ_{C} Total flux of crawl space air

Φ₁ Total flux of living-room air

 $\Phi_{\rm CL}$ Flux from crawl space to living area

 Φ_{LC} Flux from living area to crawl space

Derived Φ_{OL} Flux from outside to living area

 $\Phi_{\rm OC}$ Flux from outside to crawl space

Relations

The total flux of a compartment is the sum of all air flows entering the compartment. This is equal to the sum of all flows leaving the compartment:

$$\Phi_{\rm C} = \Phi_{\rm OC} + \Phi_{\rm LC} = \Phi_{\rm CO} + \Phi_{\rm CL} \tag{11}$$

and

$$\Phi_{L} = \Phi_{OL} + \Phi_{CL} = \Phi_{LO} + \Phi_{LC}. \tag{12}$$

Calculated quantities

The following equations show how the calculated quantities follow from the measured quantities:

$$\Phi_{\rm C} = \frac{P_{\rm C}}{C_{\rm CC}},\tag{13}$$

$$\Phi_{\rm L} = \frac{P_{\rm L}}{C_{\rm LL}},\tag{14}$$

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$$\Phi_{\rm CL} = \frac{C_{\rm CL}}{C_{\rm CC}} \Phi_{\rm L} = \frac{C_{\rm CL} P_{\rm L}}{C_{\rm CC} C_{\rm LL}}, \tag{15}$$

and

$$\Phi_{\rm LC} = \frac{C_{\rm LC}}{C_{\rm LL}} \Phi_{\rm C} = \frac{C_{\rm LC} P_{\rm C}}{C_{\rm LL} C_{\rm CC}}.$$
 (16)

Derived quantities

The following equations show how the derived quantities follow from the calculated quantities:

$$\Phi_{\rm OL} = \Phi_{\rm L} - \Phi_{\rm CL} = \frac{P_{\rm L}}{C_{\rm LL}} \left(1 - \frac{C_{\rm CL}}{C_{\rm cc}} \right),$$
(17)

$$\Phi_{\rm OC} = \Phi_{\rm C} - \Phi_{\rm LC} = \frac{P_{\rm C}}{C_{\rm CC}} \left(1 - \frac{C_{\rm LC}}{C_{\rm LL}} \right).$$
(18)

Radon

The main sources of radon (²²²Rn) are the soil and building materials. The average radon concentration in outdoor air may simply be added to the indoor concentration if decay is ignored, as is derived in the equations below. Ignoring the decay of radon is justified because the ventilation rates of the compartments investigated are generally two orders of magnitude larger than the decay rate of radon.

 $S_{\rm C}$, $S_{\rm L}$ Radon activity sources in crawl space and living area (Bq h⁻¹) $a_{\rm O}$, $a_{\rm C}$, $a_{\rm L}$ Activity concentrations outside, in crawl space and in living room (Bq m⁻³)

 $A_{\rm C}$, $A_{\rm L}$ Surplus activity concentrations:

$$A_{\rm C} = a_{\rm C} - a_{\rm O} \tag{19}$$

and

$$A_{\rm L} = a_{\rm L} - a_{\rm O}. \tag{20}$$

The radon activity sources are derived from the following balance equations (decay is ignored):

$$S_{\rm C} = (\Phi_{\rm CO} + \Phi_{\rm CL})a_{\rm C} - \Phi_{\rm OC}a_{\rm O} - \Phi_{\rm LC}a_{\rm L} = -\Phi_{\rm LC}A_{\rm L} + \Phi_{\rm C}A_{\rm C}$$
 (21)

and

$$S_{\rm L} = (\Phi_{\rm LO} + \Phi_{\rm LC}) a_{\rm L} - \Phi_{\rm OL} a_{\rm O} - \Phi_{\rm CL} a_{\rm C} = +\Phi_{\rm L} A_{\rm L} - \Phi_{\rm CL} A_{\rm C}.$$
 (22)

Solving these two equations for $A_{\rm L}$ yields

$$A_{\rm L} = \frac{\Phi_{\rm CL} S_{\rm C} + \Phi_{\rm C} S_{\rm L}}{\Phi_{\rm L} \Phi_{\rm C} - \Phi_{\rm LC} \Phi_{\rm CL}}.$$
 (23)

This equation demonstrates that the radon concentration in the living room consists of three components,

$$a_{\rm L} = a_{\rm L0} + a_{\rm L1} + a_{\rm L2} \,, \tag{24}$$

with:

$$a_{1,0} = a_{0},$$
 (25)

$$a_{L1} = \frac{\Phi_{CL} S_C}{\Phi_L \Phi_C - \Phi_{LC} \Phi_{CL}} = \frac{\Phi_{CL} \left(\Phi_C A_C - \Phi_{LC} A_L \right)}{\Phi_L \Phi_C - \Phi_{LC} \Phi_{CL}}$$
(26)

and

$$a_{\rm L2} = \frac{\Phi_{\rm C} S_{\rm L}}{\Phi_{\rm L} \Phi_{\rm C} - \Phi_{\rm LC} \Phi_{\rm CL}} = \frac{\Phi_{\rm C} \left(\Phi_{\rm L} A_{\rm L} - \Phi_{\rm CL} A_{\rm C}\right)}{\Phi_{\rm L} \Phi_{\rm C} - \Phi_{\rm LC} \Phi_{\rm CL}}.$$
 (27)

Note that

$$a_{\rm L1} + a_{\rm L2} = A_{\rm L} \,. \tag{28}$$

In the majority of the dwellings, the effective flow from the living area to the crawl space, Φ_{LC} , is almost zero. If this is the case the contribution of sources of radon in the crawl space to the radon concentration in the living room, a_{L1} , may be written as

$$a_{\rm L1} \approx \frac{\Phi_{\rm CL}}{\Phi_{\rm L}} A_{\rm C} = \frac{C_{\rm CL}}{C_{\rm CC}} A_{\rm C}, \tag{29}$$

or in words:

The part of the radon concentration in the living room that is due to a source in the crawl space, is found by multiplying the *surplus* (outside concentration subtracted) radon concentration in the crawl space by the concentration ratio (living room over crawl space) of the tracer gas released in the crawl space.