Exposure to natural radioactivity in the Netherlands

de Jong, Peter

Publication date:
2010

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):
de Jong, P. (2010). *Exposure to natural radioactivity in the Netherlands: the impact of building materials*. s.n.

5

CHAPTER 5

SUMMARY, CONCLUSIONS AND OUTLOOK

5.1 Introduction

The progeny of ^{222}Rn is one of the most extensively investigated carcinogens. Epidemiological studies of ^{222}Rn-exposed underground miners have demonstrated clearly an excess risk of lung cancer at relatively high concentrations. Extrapolation from these studies to lower exposures has suggested that, next to smoking, residential ^{222}Rn is the second leading cause of lung cancer for the general population, provided that there is no threshold for cancer induction. However, breathing rates and smoking habits of miners are not representative for the general population and, moreover, the mine and home environments differ substantially, which made these risk projections uncertain.

Combined analysis of residential case-control studies dispelled these uncertainties and determined the increase lung cancer risk at about 16% per 100 Bq m^{-3} ^{222}Rn gas (Darby *et al*., 2004). If this is approximately correct, then about 2% of the cases of lung cancer in the Netherlands can be attributed to ^{222}Rn progeny, equivalent to 200 deaths per annum. Including the exposures due to ^{220}Rn progeny and external radiation, the total number of fatalities due to natural radiation in homes is estimated to be about 650 each year, with a 95% confidence interval of 250 to 1300.

The aim of this thesis was to determine the role of building materials as part of the natural radiation exposure in the Netherlands. For that, in chapter 2 methods for determination of the ^{222}Rn exhalation rate and absorbed dose rate in air from building materials have been introduced and validated. In chapter 3 these methods, together with one for the determination of the natural radioactivity concentrations are applied to a cross section of the building materials available at the Dutch market. An investigation of the indoor radiation exposure in a small housing estate is also part of this chapter. Chapter 4 includes studies after the possibilities to reduce the resulting exposure, i.e. the retarding effect of surface coatings and the effect of composition and production on the exhalation rate from samples of concrete.

In this chapter the highlights in the chapters 2 to 4 are put forward and discussed.

5.2 Methods

Chapter 2 of this dissertation describes the development of methods that were applied to map the situation in the Netherlands. The first method concerns the determination of ^{222}Rn exhalation rates of the most common stony building materials. It is a so-called purge-and-trap method in which a sample is enclosed in a container from which the ^{222}Rn is continuously purged by a constant flow of nitrogen gas and directed through a trapping agent. The trapped activity is quantified by liquid scintillation counting (LSC).

Three performance characteristics of the method have been assessed, i.e. (a) the repeatability; (b) the intralaboratory reproducibility; and (c) the interlaboratory reproducibility. The values of these quantities range from 3 to 6% (section 2.2). These values should be considered in view of the large spread in exhalation rates from sample to sample, even within the same class of materials (Table 3.3); in that context uncertainties introduced by the measurements become insignificant compared to these inherent variations.

The limit of detection of the purge-and-trap method depends on (a) the analytical method to quantify the trapped activity; (b) the counting time; (c) the size of the analytical sample; and (d) the duration of the trapping period. Using LSC as analytical method and a counting time of 1 h, the standard conditions applied throughout this dissertation, the detection limit is calculated as 11 mBq ^{222}Rn, according to the method described by Currie (1968). For most building materials an absorption time of half an hour suffices; in special situations, for instance for bricks, longer absorption times may be necessary.

Although the method dates from the early 1990's, it is still state-of the-art; in 2001 the method was adopted as the primarily method in the Dutch standard NEN 5699 (NEN, 2001b). It has been used to verify the agreements in a covenant between the Dutch government and the Dutch building industry (De Wit and Nijland, 2009). The Working Group on radioactivity measurements of Technical Committee 85 (Nuclear Energy), Subcommittee 2 (Radiation Protection) of the International Organization for Standardization (ISO) has adopted a resolution to include the method into a new standard ISO 11665-9 (ISO/AFNOR, 2009). In view of the Construction Product Directive (EC, 1988), discussed in section 1.7, there is an urgent need for such a standard procedure.

In section 2.3 we present a method that allows the evaluation of gamma radiation doses in large groups of dwellings. The basis for these calculations is formed by a fixed set of specific absorbed dose rates, as determined for a standard geometry defined by Koblinger (1978). This quantity is defined as the absorbed dose rate in air due to an activity concentration of a construction material of 1 Bq kg^{-1} of the parent radionuclide in secular equilibrium with their daughters. Correction factors have been assessed that quantify the influence of various room and material related parameters on these specific absorbed dose rates. To verify the accuracy, our method is applied to three Dutch reference dwellings, i.e. a row house, a semi-detached house and an apartment building. The results of the model are compared with those found by MCNP (Briesmeister, 2000), a well-benchmarked multi-purpose Monte Carlo code. The differences are found to range from 3 to 7%, with an average of 4%. The advantage of our method over MCNP is that its computing time is much shorter, making it especially suited to evaluate large clusters of dwellings.

5.3 Concentration and exposures

In chapter 3 four studies have been presented on measured concentrations and exposure levels. The first study reports on a nation-wide survey, held in 2001, on the natural radioactivity concentrations and ^{222}Rn exhalation rates of the prevailing building materials in the Netherlands. The highest radionuclide concentrations were found in a porous inner wall brick to which fly ash was added. The second highest were clay bricks with average ^{226}Ra and ^{228}Ra levels around 40 Bq kg^{-1}. Concrete and mortar show the highest exhalation rates with a fairly broad range of 1 to 13 μBq kg^{-1} s^{-1}. Low natural radioactivity levels are associated with either natural gypsum or gypsum from flue gas desulphurization units, and low exhalation rates were found for clay bricks.

Natural radioactivity concentrations in the main building materials available on the Dutch market are in general on the low side of the range reported in a European-wide literature survey (EC, 1999a). As a consequence the absorbed dose rates in Dutch living rooms due to gamma radiation are also at the low side (UNSCEAR, 2000). The building material induced annual indoor effective dose is calculated at a mean and SD of 0.25 ± 0.03 mSv if 7000 h per year are spent indoors (section 3.4).

From the third national ^{222}Rn survey Bader and colleagues (2009) concluded that building materials contribute for about 70% to the indoor ^{222}Rn level in living rooms. This value is based on the difference between the ^{222}Rn efflux and influx of the living room. A similar value is obtained from the ^{222}Rn exhalation rates of the various building materials as determined in section 3.1 in combination with the average occurrence of these building materials in Dutch livings (De With and De Jong, 2009). In that study it is shown that almost 90% of the building material related ^{222}Rn originates from the concrete construction elements. From a control study carried out in 2008, it was concluded that the radiation dose due to this material has not changed with time (De Wit and Nijland, 2009).

In the second study a national survey is presented on natural radioactivity concentrations in gypsum plasters and mortars (section 3.2) and reflects the situation around 1995. The study encompassed materials from several manufacturers and included products for the non-professional sector. Three classes of material could be distinguished: class I materials of low activity concentrations, i.c. natural gypsum; class II products with intermediate levels (100-150 Bq kg^{-1} of both ^{226}Ra and ^{228}Ra); and class III products with levels up to 200-250 Bq kg^{-1} per radionuclide. The elevated levels of ^{226}Ra and ^{228}Ra as found in gypsum plasters will, in addition, lead to enhanced gamma dose rates and ^{222}Rn concentrations in homes. Nevertheless, in view of the thin layers of plasters compared to the total building mass of a dwelling, these increases will be negligible. A high ^{228}Ra-activity concentration in plaster will however result in an increase of the ^{220}Rn exhalation rate and hence to elevated indoor ^{220}Rn progeny levels and lung doses. A worst-case estimate suggests an extra residential effective dose of around 2 mSv per annum (De With and De Jong, 2009).

The third study (section 3.3) deals with exposure to ^{222}Rn and external radiation of inhabitants in a new housing estate. In this estate 101 houses were built at about the same time, but according to nine designs. The effective dose to inhabitants ranged from 0.4 to 1.4 mSv per year, depending on the design. The houses at the low end of this range are characterized by a timber frame construction with an airtight concrete ground floor, with the remaining floors made of timber and with flue gas desulphurization gypsum inner walls; the radiation burden of the inhabitants resembles the outdoor situation. Designs constructed according to a ‘conventional’ building method (concrete floors, cavity walls made of brick and sand-lime brick), show similar ^{222}Rn concentrations and dose rates. One of the main findings of this study is that the design has a more pronounced influence on the radiation burden than habits and preferences of its occupants. A similar conclusion can be drawn from the results published by Lomas and Green (1994), who found no significant difference in ^{222}Rn concentration after new occupants moved into a dwelling.

The distribution of gamma dose rates due to building materials in the Netherlands is the topic of the fourth study (section 3.4). The model described in section 2.3 is applied to a representative set of dwellings of which the areas occupied by the various building material are well documented. Using a Monte Carlo method, building material and housing data are combined with activity concentrations and densities of 90 samples of building material, as emerged from section 3.1. By simulating 100,000 combinations, the distribution of the absorbed dose rate in Dutch livings is computed and found to be near Gaussian with an average and SD of 51 ± 6 nGy h^{-1}. After correction for the indoor contribution of cosmic and terrestrial radiation, their actual distribution corresponds to within 5% with measurements.

The last study is particularly relevant with respect to a recent publication of the European Commission on the revision of the Basic Safety Standards (EC, 2009). The proposal involves a limitation of the gamma dose rate induced by building materials, according to a simplified calculation model presented in section 1.7. In this EC-model no corrections factors are applied for the differences between dwellings, leading to a severe overestimation of the building materials induced dose rates. The model described in this dissertation takes almost the complete set of parameters into account. Although not all considered parameters are equally important, it clearly demonstrates the shortcomings of the European Commission’s model and, to a lesser extent, that of the national model, known as the Stralingsprestatienorm (NEN, 2002).

5.4 Radon-transport mechanisms and mitigation

In the Netherlands building materials account for about 70% of the indoor ^{222}Rn concentration and hence various ways have been investigated to reduce their ^{222}Rn release rates. Knowledge of the underlying mechanisms is essential, because it may lead to new or additional reduction possibilities. The application of surface coatings to reduce ^{222}Rn exhalation rates in an existing situation has been verified by a number of investigators. The study in section 4.1 has evaluated the retaining effect of over 20 surface coatings commonly used in the Netherlands for decoration of walls and ceilings. These coatings were found to be ineffective. Reductions up to 75% can be achieved by industrial surface coatings based on epoxy resin and polyurethane, when applied to all sides of test walls. According to model calculations, the exhalation rates could be reduced by 95% if these coatings were applied to the inward facing surface of walls only. Lesions, however, can negate a large portion of the sealing effectiveness and the same applies for cracks that may develop over time in the paint layer due to ageing.

Concrete is found to be a major contributor to indoor ^{222}Rn. In the construction of houses various kinds of concretes mixtures are applied. In the sections 4.2 and 4.3 the influence of several parameters on the ^{222}Rn exhalation rate has been investigated. In section 4.2 the parameters related to composition and production processes on the ^{222}Rn exhalation rate of concretes were studied. Especially the water evaporation rate during conditioning was found to have a strong positive correlation with the calculated ^{222}Rn emanating power.

Within a series of 23 concrete mixtures, concrete compositions have been varied by changing the amount of cement, type of cement (Portland or blast furnace slag cement), water-to-cement ratio, use of an air entraining agent or the amount of recycled granulates. This resulted in a range of porosities from 1% to 16% (section 4.3). The ^{222}Rn exhalation rate is normalized to the ^{226}Ra level and expressed in the so-called ^{222}Rn release factor. Most ^{222}Rn originates from the cement and therefore a second ^{222}Rn release factor is based on the amount of ^{226}Ra in the cement fraction. Although the methods to attain the porosities in the concrete mixtures differ widely, this cement-related factor corresponds well with the capillary porosity of the mixtures. Since the water-to-cement ratio of the fresh paste is a good indicator of the capillary porosity, this is the guiding factor in the fabrication of concretes low in ^{222}Rn exhalation. The lower the water-to-cement ratio, the less capillary pore area will be available from which ^{222}Rn can emanate from the mineral matrix into the pore system. The good correlation between the cement-based ^{222}Rn release factor and literature data on the internal capillary pore area support the results of this study.

5.5 Final remarks and outlook

Building materials always have received special attention from the Dutch government, illustrated by the number of studies that were commissioned to develop a method to calculate and control building material-induced exposure of inhabitants (Ackers, 1990; Van Heijningen and Ackers, 1990; VROM, 1990b; Roelofs and Wiegers, 1995; NEN, 2002). According to the latest national ^{222}Rn survey, covering 1000 newly built houses in the period 1994-2003, the average ^{222}Rn concentration in the living room is 13-14 Bq m^{-3} (Blaauboer *et al.*, 2007). As outlined in Table 5.1 the Netherlands has the lowest average ^{222}Rn level in the European Communities. The main four reasons are:

- a low ^{238}U contents of Dutch soils;
- a low ^{222}Rn release due to a high groundwater level in large parts of the country;
- a common building practice with well-ventilated crawl spaces; and
- a legal maximum air permeability of the ground floor.

Table 5.1
Average ^{222}Rn concentrations in dwellings in some European countries (UNSCEAR, 2009).

Country	**^{222}Rn (Bq m^{-3})**
The Netherlands	14[a]
United Kingdom	20
Belgium	48
Germany	50
Denmark	59
France	62
Spain	90
Sweden	108
Finland	120

[a] Preliminary value (Blaauboer *et al.*, 2007).

Therefore, unlike most other countries, the soil beneath the houses only plays a minor role as a source for indoor ^{222}Rn. Consequently, building materials play a more prominent one and are estimated to be responsible for 70% of the indoor ^{222}Rn concentration. The contribution of building materials to the external exposure in houses is calculated at 55% (Table 1.8), leading to an average total exposure due to building materials of 0.6 mSv per year. This corresponds to about 40% of the average radiation burden due to natural radiation sources in the Netherlands (Van Bruggen *et al.*, 2004). In summary it may be stated that the situation in the Netherlands with respect to ^{222}Rn and external radiation in dwellings is reasonably well understood.

Things are different for ^{220}Rn. Thus far, ^{220}Rn or its short-lived decay products have not been investigated for dwellings and building materials in the Netherlands. Therefore the number of potential fatalities due to exposure to ^{220}Rn progeny, calculated as 60 per year, is based on model calculations as reported by De With and De Jong (2009). In that study it was assumed that building materials were not covered with a plaster layer or paint coating. As suggested in section 3.2 and confirmed by model calculations, application of plasters high in ^{228}Ra can seriously enhance the inhabitants' radiation burden and consequently the number of fatalities.

As indicated in section 1.8.2 the average ^{222}Rn concentration as found in the third national survey is more than a factor of two lower than expected on the basis of the results of the second survey. As reported by Vargas and Ortega (2007) the ^{222}Rn measuring devices applied in the second survey show an unintended high sensitivity for ^{220}Rn of about 50%. As the difference between the results of both surveys is of the order of 15 Bq m^{-3}, the average ^{220}Rn concentration near the measuring devices would be about 30 Bq m^{-3}. It is, however, not really possible to reconstruct an (average) source term for ^{220}Rn from this finding. Due to the short half-life of ^{220}Rn, there is a steep concentration gradient of the ^{220}Rn concentration from the surface of construction elements to the centre of a room. The shape of this curve is seriously affected by the so-called turbulent diffusion coefficient (De With and De Jong, 2009), an indicator for mixing of indoor air. This parameter is mainly influenced by time-dependent temperature-driven airflows and is not well known. Conducting a survey on ^{220}Rn concentrations in Dutch dwellings to obtain a better insight in the ^{220}Rn source term in the radiation burden is therefore only meaningful if such a study is combined with explicit attention for this parameter.

Another way to get knowledge on the ^{220}Rn source term, and with that on the exposure of the Dutch population to short-lived decay products of ^{220}Rn is to survey ^{220}Rn exhalation rates of Dutch building materials, comparable with that on ^{222}Rn as reported in section 3.1. Since practically all ^{220}Rn originates from building materials, the source term can be obtained by combination of exhalation rate data with results on the distribution of the various building materials in Dutch dwellings. The exhalation rate of plaster should be considered in this survey, since, as indicated above this material may have high exhalation rates. Determination of retarding effects by coatings and wallpapers regularly applied in Dutch households, will be part of such a study. Although reported in section 4.1 that these materials have no effect with regard to the exhalation of ^{222}Rn, due to the factor 75 lower ^{220}Rn diffusion length, there might be a significant reduction of ^{220}Rn exhalation.

Aim of the suggested studies is to attain a similar knowledge base for ^{220}Rn as for ^{222}Rn, but is experimentally a great challenge.