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INTEGRATED CRITERIA DOCUMENT RADON

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Integrated Criteria Document Radon

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dispersion, concentrations in the environment, exposures, human toxicity, radon control techniques, costs of radon control, economic consequences, risk analysis, models

Summary

This document contains a systematic review and a critical evaluation of the most relevant data on the priority substance radon for the purpose of the effect-oriented

environmental policy

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Lay-out

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This integrated criteria document deals with the element radon and its short-lived daughter products. When the term "radon" is used, it refers to all the isotopes of this element. Most information concerns the relatively long-lived 222 Rn ($t_{1/2}=3.8$ days). In addition, some information on the short-lived 220 Rn ($t_{1/2}=56$ seconds) has also been included. The historical term "thoron" for 220 Rn has not been used in this document.

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SUMMARY

This document on the subject of radon contains data on sources, emissions, dispersion and risks. Risks are based on a comparison of exposure levels and detrimental effects. Radon is a chemically inert, naturally occurring radioactive gaseous element. The main risk to humans from radon is the induction of lung cancer. In this context, the short-lived daughter products of ²²²Rn and ²²⁰Rn are of special importance. The risks to plants and animals, in terms of possible extinction of species, are negligible. In view of the nature of the risk of radon and its occurrence, attention has been paid chiefly to the risks to humans in the indoor environment. Possible techniques to reduce these risks, as well as the costs involved, are discussed. A number of potential policy scenarios for radon are also examined.

Although various sources contribute to radon emission, all sources are derived directly or indirectly from the soil. The total annual emission in the Netherlands is about $2x10^{16}$ Bq of 222 Rn and about $1.5x10^{18}$ Bq of 220 Rn. The 222 Rn emission is determined primarily by the soil itself (about 95% of the total emission). Consequently, the contribution of other sources, such as ore tailings, phosphate fertilizers and building materials, to the 222 Rn concentration in the outdoor atmosphere is small.

Because of the short half-life of ²²⁰Rn (56 seconds) in comparison with ²²²Rn (3.8 days), ²²²Rn is most significant for human exposure, so that the emphasis in this integrated criteria document is on ²²²Rn. The exposure received indoors is much greater than the outdoor exposure due to the accumulation of ²²²Rn inside buildings, combined with the relatively long periods of time most individuals spend indoors. The soil (averaging about 70%) and the building materials used (about 30%) are the principal contributors to the ²²²Rn concentrations in indoor air. The contribution of the other sources to the ²²²Rn concentration is about 1% on average. The mean ²²²Rn concentration in Dutch living rooms is 29 Bq m⁻³ (range 8-140 Bq m⁻³), which falls within the range of 20-90 Bq m⁻³ observed in other European countries. Approximately 0.7% of the living rooms have ²²²Rn levels of 100 Bq m⁻³ or above, which means that the exposure received by about 100,000 persons is at least three times greater than the average. The mean outdoor atmospheric concentration in the Netherlands is about 3 Bq m⁻³ (1-10 Bq m⁻³, depending on the geographical location).

Reliable measurement techniques are available for measuring radon in air as well as in other matrices (building materials, soil, drinking water). Also, static and dynamic models have been developed for estimating radon concentrations inside houses and for determining the effect of control measures. Validation of these models by means of field tests is desirable.

The main risk from exposure to radon and radon daughters is the induction of lung cancer, with lung cancer mortality being used as the risk criterion. As the average dose conversion coefficient (effective dose equivalent per unit exposure), a value of 0.14±0.05 mSv a⁻¹ per Bq m⁻³ (EEC) (10±4 mSv WLM⁻¹) is used for ²²²Rn. For ²²⁰Rn, this conversion coefficient is 0.6±0.2 mSv a⁻¹ per Bq m⁻³ (EEC) (3±1 mSv WLM⁻¹). The lifetime risk of lung cancer mortality due to a lifetime exposure to ²²²Rn and progeny is estimated to be 350x10⁻⁶ WLM⁻¹, based on epidemiological data from (uranium)miners, and assuming a linear dose-effect relationship without threshold. For ²²⁰Rn and progeny,

the lifetime risk is 150x10⁻⁶ WLM⁻¹, using the results from dosimetric models and the ICRP conversion factor from dose to risk. The uncertainty in the risk values is about a factor of two. Based on these risk values, the average exposure to radon daughters in the Netherlands corresponds to a lung cancer mortality of 60 (uncertainty interval: 30-120) per million persons per year (80% resulting from ²²²Rn and 20% from ²²⁰Rn). These risks are controllable only to a limited extent and do not fall under the risk limits proposed by the radiation protection policy.

There are indications that ²²²Rn can cause cancers other than lung cancer, for example, leukaemia and mammary carcinomas. Appropriate experimental research with laboratory animals is recommended.

If no control measures are taken, the average risk for members of the population will increase as a result of the construction of new dwellings, which have higher average ²²²Rn concentrations than existing dwellings. With the current policy on building materials, aimed at ensuring that the radiation risk due to building materials to occupants of future dwellings will not become greater than the risk incurred in existing, representative dwellings, the risk continues to increase. A future policy could concentrate on control measures in all dwellings to be constructed. This may result in an annual reduction of 0.6% in the average risk. On the basis of this document, a reduction objective will be formulated.

Calculations using a model indicate that an increased ventilation rate in the crawl space, combined with a well-sealed floor slab, appears to be the most cost-effective: for future housing, the resulting individual dose reduction of the occupants is 0.4 mSv per year on average, at an increase in the construction costs of Dfl. 500.- to Dfl. 2000.- per dwelling. The annual costs are estimated to be Dfl. 70.- to Dfl. 170.-. It is expected that these costs will not significantly affect the volume of the building activities.

The risks from radon are due partly to natural environmental radiation (uncontrollable), and partly to the building materials used and construction practices (controllable to a limited extent). Reducing the current risks associated with radon is desirable in view of the general aim of risk reduction. However, the possibilities for reducing the current risks are limited, while the effectiveness of the control measures with respect to reduction of the average risk is low in the short term. Attention should be focused both on the implementation of the above measures in future housing and on the development of methods for identifying houses and other buildings with relatively high ²²²Rn levels. Control measures in new construction can, in the long term, lead to a substantial reduction in both the individual risk to the occupants and the average risk. Remedial action in high-risk houses reduces especially the individual risk to the occupants. The effect of this action on the average risk from radon in the Netherlands will be marginal.

Knowledge of radon in the Netherlands is still incomplete. Research into ²²²Rn exposures in office buildings, factories, schools and day nurseries, and additional field test of the effectiveness of radon control measures are recommended. As knowledge of ²²⁰Rn exposures is virtually non-existent, exploratory research is also recommended.

INTRODUCTION

In the Netherlands, environmental policy at government level is first of all aimed at preventing or mitigating undesirable effects of human activities and practices which may affect the health of humans, animals, plants or ecosystems. These effects can occur on different spatial scales: local, regional, fluvial, continental and global (National Environmental Policy Plan, 1990-1994). In addition, environmental policy is aimed at sustainable development, which will meet the needs of today without jeopardizing future generations. However, with insufficient knowledge it is impossible for the time being to describe fully the general environmental quality desired. Attention is therefore being concentrated on factors which are believed to entail considerable risks, such as environmentally harmful substances. A selection has been made and a priority list compiled of the many substances of relevance, because of their emission or use. These are substances on which the policy theme "dispersion" primarily concentrates. So-called integrated criteria documents are drawn up for most of these priority substances. Radon is on the list of priority substances for the period 1990-1994 (National Environmental Policy Plan, NMP). The principle of risk control presented in the NMP has been further elaborated in the policy paper "Radiation protection and risk management" (1990).

Integrated criteria documents contain, per substance or substance group, data on the sources and the distribution pattern (soil, water, air, biota), the risks of current and expected exposure concentrations for humans, (parts of) ecosystems and materials. Assembling all relevant information on a specific substance, for the purpose of establishing environmental quality criteria and (if desired) source-oriented criteria, forms part of the objective of integrated criteria documents. The objective also includes indicating the technical possibilities and the economic consequences of risk reduction on the basis of a number of policy-relevant scenarios. This information serves as the scientific basis for formulating the effect-oriented environmental policy. This policy is aimed at attaining as large a risk reduction as possible, the desirable level being the ultimate goal. This level is the concentration in the environment at which no adverse effects (i.e. the risks of which are considered to be negligible) occur for humans, animals, plants, ecosystems and materials. If the desirable level cannot be reached within a reasonable period of time, a limit value is established for a limited period, in which the risks lie between the maximum permissible and the desirable concentration. In addition to the possible risk reduction, economic and social factors are also determinants in setting this limit value. This document is confined to the provision of information necessary for the establishment of the above-mentioned environmental quality criteria in terms of policy; the information supplied may also result in a general task-setting for the emission reductions per source type. The sections of this document do not, therefore, contain a policy opinion.

The National Institute of Public Health and Environmental Protection (RIVM) is responsible for drawing up integrated criteria documents. The Radiological Service of the TNO (RD-TNO*) and the Institute for Applied Radiobiology and Immunology (ITRI-TNO) of the Division of Health Research of the TNO participated in the realization of this report.

(*) The RD-TNO has been integrated with the ITRI-TNO since 1-1-1991, but operates under its original name.

Government, trade and industry, and representatives from scientific institutes were involved in the preparation of this document. For example, the document has been checked in its entirety by a Review Committee of the RIVM, while an Advisory Board composed of staff from the Ministry of Housing, Physical Planning and the Environment (VROM), the Ministry of Welfare, Health and Cultural Affairs (WVC), and the Ministry of Economic Affairs (EZ) gave guidance in its compilation. Trade and industry were involved through the ad hoc Working Group on Integrated Criteria Documents of the Office of the Environment and Physical Planning of the Organizations Dutch Employers' VNO and NCW.

If there are differences of opinion, an addendum drawn up by the ad hoc Working Group may be added to the document. This possibility also exists for environmental groups through the Nature and Environment Foundation (Stichting Natuur en Milieu). At a later stage, the Health Council will publish a brief report on the document, including any addenda.

This document deals with radon. Radon is a chemically inert, radioactive gaseous element of which several isotopes are known. Regarding radioactivity, only ²²²Rn and ²²⁰Rn are of concern because of their occurrence and half-lives. The short-lived daughter products of radon constitute a radiation hazard in indoor environments. Therefore, the emphasis in this document is on indoor radon, with attention being paid to measurement techniques, sources, dispersion and occurrence, as well as toxicity and the risks to humans. Only scant attention has been given to exposure of humans out-of-doors, bioaccumulation of long-lived radon daughters and risks to ecosystems.

In compliance with the wishes of the commissioning body, this report is based as much as possible on existing recent reviews; the original literature is only consulted when the review papers contain inconsistent data or conclusions, and when specific data are used for deriving the recommended levels. In addition, optimum use has been made of the information available within the framework of the research programme "Controllable forms of natural background radiation" (RENA). Substantial attention has been paid to the policy plans as laid down in "Radiation protection and risk management" (ORS). This concerns especially the controllability of a particular source, whether or not the risk limits are applicable, special attention to the issue of "Building construction and occupancy", and the position of radon in relation to the proposed risk limits for non-nuclear industries.

Scope

The Integrated Criteria Document Radon deals with the risks to man and animals from exposure to radon and its short-lived decay products. As regards the exposure to man from airborne radon (daughters), the potential routes of entry into the body are inhalation and absorption through the skin. Exposure also occurs through ingestion (food and drinking water).

- (*) It is true that food contains short-lived decay products of radon, but they do not not occur isolated from the element radium, ²²⁶Ra and ²²⁸Ra being the relevant parent nuclides. These exposures are not discussed here because the radium issue was not part of this project.
- (**) The risks from ingestion of ²²²Rn dissolved in water are an order of magnitude lower than those due to inhalation of ²²²Rn daughters arising from the release of ²²²Rn from domestic water use (see 6.1.3).

Inhalation is by far the most important exposure route (see 6.1.3), so that much attention is paid to the occurrence of radon in air, with the emphasis on the indoor environment, because of the relatively long residence times and high concentrations indoors.

Radon is a naturally occurring radioactive element. However, elevated exposures can can result from human activities. Thus, attention is paid to the emission of radon and short-lived daughters from non-nuclear industries. Also, in connection with the issue of "Building construction and occupancy", the risks from external irradiation due to short-lived radon daughters contained in building materials are evaluated, and for the sake of completeness, terrestrial radiation arising from short-lived radon daughters is briefly considered. The risks from ²¹⁰Pb abd ²¹⁰Po formed by decay of atmospheric ²²²Rn are only briefly discussed, and result from the dispersion of ²²²Rn on a global scale. The risks from ²¹⁰Pb and ²¹⁰Po discharged by non-nuclear industries were not part of this project.

Reading guide

The radon issue is complex and, because of the radioactive properties of the element, radon differs from the substances dealt with so far in the series of integrated criteria documents. Therefore, this integrated criteria document can be less accessible to the uninitiated reader. To increase its accessibility, a brief document overview and a table indicating the extent to which the nature of the information in each chapter is general or related to the Dutch situation.

This integrated criteria document is structured as follows:

The document consists of 10 chapters. Chapter 1 gives general information on the properties of the element radon and a brief description of the existing standards and guidelines. Chapter 2 deals with the sources and gives an estimate of the ²²²Rn and ²²⁰Rn emissions in the Netherlands. A general desription of the dispersion of radon in the atmosphere and the deposition of radon daughters is given in Chapter 3. This chapter also discusses the behaviour of radon in the soil and its entry into dwellings.

The measurement techniques for ²²²Rn and ²²⁰Rn in the various environmental compartments, including the important compartment "air", are presented in Chapter 4. The concentrations observed in the measurement programmes are described in Chapter 5. Chapter 6 includes general information on human toxicity, with the emphasis on dosimetric models and the conversion coefficients from exposure to risk derived from these models. Chapter 7 describes human risks and the results of applying the risk coefficients for radon to the exposure of the Dutch population.

Chapter 8 gives a general description of a physical model, specifically developed for the Dutch situation, for the entry of radon into and its behaviour inside a dwelling. This description is included because virtually all control measures proposed for the Netherlands are based on model results. In addition, the control measures which can be applied in the Netherlands are catalogued and their cost-effectiveness is indicated, and a number of policy-relevant scenarios are presented. Chapter 9 gives an estimate of the economic consequences of measures for reducing the radon exposure in the Netherlands. Finally, Chapter 10 evaluates the information in this integrated criteria document, and gives the main conclusions and recommendations.

Furthermore, it is important for the report's accessibility that each chapter is provided with a section "Summary and conclusions". This part of the text, together with the table

of contents, is pre-eminently suitable for acquainting the reader quickly with the subject matter of the chapter in question.

<u>Table</u> Summary of the structure and nature of the information in the Integrated Criteria Document Radon

Chapter	Contents	Nature of the information
1	Properties; standards	General; Dutch and international Standards and guidelines
2	Sources and emissions	General; emission relating to the Netherlands
3	Behaviour and dispersion	General
4	Measurement techniques	General
4 5 6	Concentrations and exposure	Description of the Dutch situation
	Effects	General
7	Risks	General; risks to the Dutch population
8	Modelling and measures	Emphasis on the Netherlands
9	Economic consequences	Emphasis on the Netherlands
10	Evaluation	Emphasis on the Netherlands

1 PROPERTIES, FORMATION AND EXISTING STANDARDS

1.1. PROPERTIES

The element radon was discovered by Dorn in 1900, who called it radium emanation because the gas is released from radium-bearing materials (such as virtually all soil types, various building materials, etc.). In 1908 the element was isolated by Ramsay and Gray, who named it niton (from the Latin "nitens", shining). They subsequently determined the density of this element, the heaviest of the noble gases. Radon (Rn) is a radioactive element of which 26 isotopes are known. The best known isotopes are ²¹⁹Rn (actinon), ²²⁰Rn (thoron) and ²²²Rn (confusingly often also called "radon"), which all occur in nature (see 1.2). To prevent misunderstandings, the term radon will be used hereafter only when it concerns the element. The various isotopes will always be indicated with the symbol "Rn" plus associated mass number.

Radon-222 was used in the past in the treatment of various disorders. To this end, the ²²²Rn was drawn off at a radium source and put in small tubes. Another type of "radon therapy" is practised in the so-called radon baths in, for instance, Austria, Germany, France, Italy, the United States and the former Soviet Union.

1.1.1 A few physicochemical basic data

Radon is the element with atomic number 86. The known isotopes of radon have mass numbers of 201 to 226. The best known radon isotope is ²²²Rn. Radon is colourless at room temperature, but exhibits yellow phosphorescence at temperatures below 0°C, turning to orange-red at temperatures below -200°C. The melting point and boiling point of ²²²Rn are -71°C and -62°C, respectively. Radon is a noble gas, as are helium, neon, argon, krypton and xenon. The solubility of the gas in water is low (Table 1.1). Its solubility in human fat is however relatively high compared with that in water and blood. Owing to its large atomic diameter, radon can be readily adsorbed on, for example, activated charcoal and silica gel (Weast et al., 1989).

<u>Table 1.1.</u> Solubility of ²²²Rn in water, human blood and body fat (Cothern and Smith, 1987)

Material	Temperature	Maximum ²²² Rn concentration (per Bq m ⁻³ of ²²² Rn in air) Bq m ⁻³
	°C	Bq m-3
Water	0	0.51
	20	0.25
	30	0.20
	37	0.17
	50	0.14
	100	0.11
Human blood	37	0.43
Human fat	37	6.33

Radon has a density of 9.73 kg m⁻³ (0°C, 10⁵ Pa). The density in the solid and liquid phases is 4400 kg m⁻³ and 4000 kg m⁻³, respectively.

1.1.2 Transport of radon by diffusion and convection

Because radon, as a noble gas, is chemically inert, the manner in which it is dispersed is particularly important with regard to human exposure. Radon can be transported through soil, but also, for example, through ores and building materials in two ways, namely, by diffusion and convection.

Diffusive transport

Diffusion plays an important role in the release of radon from soil, building materials, ores, etc. This process is described by Fick's law:

$$\Phi = -D_{\epsilon} \cdot \nabla A_{R_{n}} \tag{1.1}$$

where Φ (Bq m⁻² s⁻¹) is the radon flux through a unit area of the bulk material of the soil or building material, D_e (m² s⁻¹) is the effective bulk diffusion coefficient, and ∇A_{Rn} (Bq m⁻⁴) is the activity concentration gradient of radon in the pore space.

Various models for this diffusion process have been proposed in the literature (e.g. Buckingham, 1904; Penman, 1940; Currie, 1960). In most models, porosity (the fraction of the soil or building material filled with water and air) and the shape of the particles, from which radon escapes, play a role. The pore size in the source material is important for the diffusion process. Diffusion occurs primarily through the large pores. Typical values for permeability (see Convective transport), porosity and particle size are presented in Table 1.2 for different soil types, as are values for the diffusion coefficient derived for these soils. In general, it can be stated that the diffusion coefficient is of the order of 10⁶ m² s⁻¹ for a soil with a moderate moisture content (about 50% of the porosity) and that it decreases to about 2x10⁻¹⁰ m² s⁻¹ for saturated soils. The low diffusion rate in saturated soils is caused mainly by the low diffusion rate in water compared with that in air (D is about $1x10^{-9}$ m² s⁻¹ and $1.2x10^{-5}$ m² s⁻¹, respectively) (see also 1.2.3). Building materials exhibit a similar range. Diffusion coefficients for radon in building materials of 6.8x10¹⁰ m² s⁻¹ (heavy concrete) to 1.7x10⁻⁶ m² s⁻¹ (gypsum) have been measured (Folkerts et al., 1984). The diffusion coefficient is highly dependent on the porosity and permeability of the material concerned.

Table 1.2. Typical values for permeability, porosity and effective bulk diffusion coefficients of a few soil types

Soil description	Permeability	Porosity	Moisture content	Diffusion coefficien	Particle t size
	1)	2)	3)		5)
	m ²	€ ′	w	4) m² s ⁻¹	μ m
Dune sand	10-10	0.45	0.16	4.2x10 ⁻⁶	50 - 420
Windborne sand deposi	t 5x10 ⁻¹²	0.40	0.43	1.6x10 ⁻⁶	50 ~ 420
Marine clay loam	10 ⁻¹⁵	0.50	0.66	5.9x10 ⁻⁷	< 210
River clay	10-16	0.55	0.89	6.3x10 ⁻⁸	< 150

¹⁾ Nazaroff and Nero, 1988

²⁾ fraction of the soil volume occupied by pores

³⁾ moisture content at field capacity (water has drained from saturated soil) as a fraction of the pore volume (Kuipers, 1981) 4) fitted function: $D_e = 7 \times 10^{-6} \exp[-4(w - w.\epsilon^2 + w^5)]$ (Rogers et al., 1984)

⁵⁾ Kuipers, 1981

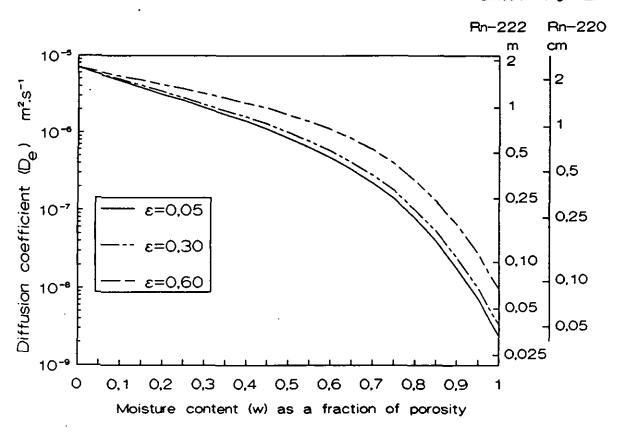


Figure 1.1. Relationship between diffusion length, diffusion coefficient, porosity and moisture content using the formula of Rogers et al. (1984) and that of the diffusion length.

The diffusion length L can now be defined as the average distance traversed due to diffusion of a radioisotope in a given material within one half-life of the isotope under consideration, or:

$$L^2 = \underline{D}_c \tag{1.2}$$

where D_e is the diffusion coefficient in m^2 s⁻¹ and λ is the decay constant of the isotope under consideration (see 1.2) in s⁻¹. Figure 1.1 shows the relationship between diffusion coefficient, diffusion length, porosity and moisture content, as determined with the aforementioned formula of Rogers et al. (1984) and the definition of the diffusion length. As the moisture content increases, the diffusion coefficient drops slowly at first, when the small pores fill up. However, when the larger pores also begin to fill with water, the diffusion coefficient drops much more rapidly ($w \ge 0.6$). For a given moisture content, the number of air-filled pores increases with increasing porosity, resulting in a greater diffusion coefficient. When the moisture content is practically zero, however, recoil losses occur (see 1.2.3) and the porosity is much less important.

A sample calculation: a common (see Figure 1.1) diffusion length (= average diffusion distance traversed within one ²²²Rn half-life) of 1 m and a free ²²²Rn concentration in soil (in the pores) of 50,000 Bq m⁻³ would, with an infinitely thick homogeneous soil layer and

an infinite volume of air above it, result in a 222Rn exhalation rate of:

$$\frac{50,000 \text{ Bq m}^{-3} \cdot 1 \text{ m}}{3.8 \text{ d} \cdot 24 \text{ h} \text{ d}^{-1} \cdot 3600 \text{ s} \text{ h}^{-1}} \approx 0.15 \text{ Bq m}^{-2} \text{ s}^{-1}$$

Convective transport

The air permeability of the source material determines to a considerable extent whether convection or diffusion is the dominant transport process. For coarse-grained sandy soils, convection usually dominates, and diffusion is important for clay (small particle size). If the soil permeability is constant and isotropic, the rate of convective transport follows from Darcy's law:

$$v = -\frac{k}{\mu} \nabla P \tag{1.3}$$

where v (m s⁻¹) is the net convective velocity, k (m²) is the intrinsic permeability, μ (Pa . s) is the viscosity of air (1.71 x 10⁻⁵ Pa s at 0°C), and ∇P (Pa m⁻¹) is the pressure gradient. A certain underpressure above the soil or on one side of a building material induces convective transport of radon. For a typical pressure difference of 5 Pa and a permeability of 10^{-11} m², this will, according to equation (1.3), result in a net transport rate of about 3 μ m s⁻¹ from the soil or building material. This rate seems slow, but multiplying it by a free ²²²Rn concentration in soil of 50,000 Bq m⁻³ gives an exhalation rate of 0.15 Bq m⁻² s⁻¹.

As a measure of the relative importance of convective transport with respect to diffusive transport, the parameter P* (a Péclet number) has been defined (Nazaroff and Nero, 1988):

$$P^{\bullet} = \underbrace{k \in \Delta P}_{\mu \ D_{c}} \tag{1.4}$$

For a pressure difference ΔP of 5 Pa, a porosity ϵ of 0.5 and a diffusion coefficient D_{ϵ} of 10^{-6} m² s⁻¹, these two modes of transport will be of equal importance ($P^*=1$) when the permeability is about $7x10^{-12}$ m². Convection is the dominant transport process when P^* is much greater than 1 and diffusion when P^* is much smaller than 1.

1.2 FORMATION

The radioactivity of a nuclide arises from the decay of the nucleus of this nuclide. Thus, the radon isotopes considered here are the decay products of radioactive isotopes of the element radium. They decay with the emission of particles and/or radiation. Ionizing radiation is of three types: alpha (helium nuclei), beta (electrons) and electromagnetic (photons: gamma radiation from the nucleus, and X-radiation from the electron cloud surrounding the nucleus) radiation. In the decay of radon, it is the alpha-emitting decay products which are of most significance for human exposure.

1.2.1 Radioactive decay

The activity A of N atoms of a radioactive nuclide means the number of "nuclear

transitions" per second, or the decrease in the number of N atoms per second:

$$A(t) = -\frac{dN(t)}{dt}$$
 (1.5)

with the unit being the becquerel (1 Bq = 1 s⁻¹). With regard to radioactive decay of a nuclide, the rate of decrease of the number of nuclides is proportional to the number of nuclides present (N(t)):

$$\frac{dN(t)}{dt} = -\lambda N(t) \tag{1.6}$$

where λ is defined as the decay constant of the nuclide concerned (s⁻¹). The solution to this differential equation is:

$$N(t) = N_o e^{-\lambda t} ag{1.7}$$

where

$$\lambda = \frac{\ln 2}{t_{1/2}} \tag{1.8}$$

 N_o is the number of nuclides at t=0, and $t_{1/2}$ is the half-life of the isotope concerned (the time in which the activity and the number of nuclides decrease by 50%. Combining the equations (1.5), (1.6) and (1.7) yields for the activity of the isotope concerned:

$$A(t) = A_o e^{-\lambda . t}$$
 (1.9)

where A_o is the initial activity λ N_o . When the decay product (also called daughter product) is also radioactive, we speak of a radioactive series. Then:

$$\frac{d\mathbf{N}_1(t)}{dt} = -\lambda_1 \ \mathbf{N}_1(t) \tag{1.10a}$$

$$\frac{dN_2(t)}{dt} = +\lambda_1 N_1(t) -\lambda_2 N_2(t)$$
(1.10b)

etc.

If in such a series the daughter product (N_2 in equation 1.10b) decays faster (shorter half-live) than the mother nuclide, the daughter "grows in" till it has the activity of the mother nuclide. Such a situation is called radioactive equilibrium.

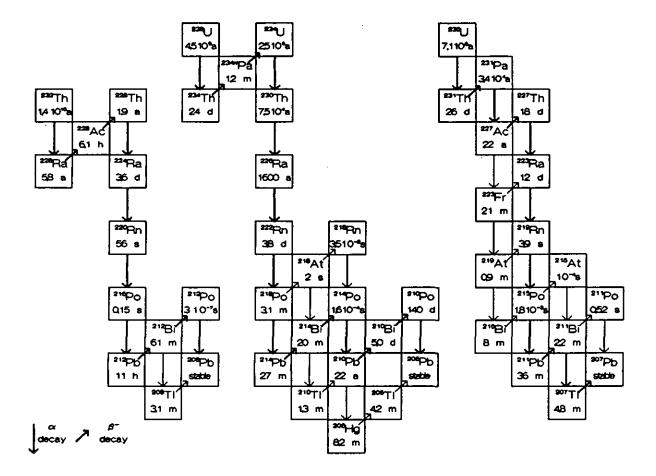


Figure 1.2. Decay series of the primordial radionuclides ²³⁵U, ²³²Th and ²³⁸U; the half-lives and the alpha and beta decay are shown (heavy arrows indicate the most important transitions).

1.2.2 Decay series of primordial radionuclides

The noble gas radon is a radioactive decay product of radium. The best known isotopes ²¹⁹Rn, ²²⁰Rn and ²²²Rn are decay products in the decay series of ²³⁵U, ²³²Th and ²³⁸U, respectively (see Figure 1.2), also called primordial radionuclides. The type of radiation (alpha, beta) and the half-lives of the various nuclides in the uranium and thorium series are shown in this figure. All the nuclides of these series occur in soil. The average concentrations of the three heads of these series, ²³²Th, ²³⁵U and ²³⁸U, in soil are 25 Bq kg⁻¹, 1.1 Bq kg⁻¹ and 25 Bq kg⁻¹, respectively (UNSCEAR, 1988). The radon isotopes of these series are present at approximately the same concentrations.

Radon and radon decay products

The radon isotopes ²¹⁹Rn and ²²⁰Rn decay through short-lived isotopes of polonium, bismuth and lead to stable isotopes of lead. During the decay of ²²²Rn, two longer-lived nuclides (²¹⁰Pb and ²¹⁰Po) are formed (see Figure 1.3). All three radon isotopes will diffuse from the soil into the atmosphere. Since the natural abundance of ²³⁵U and thus also of ²²³Ra (see Figure 1.2) is much lower than that of members of the ²³⁸U and ²³²Th

decay series, and since the half-life of ²¹⁹Rn is also much shorter than that of the other two radon isotopes, substantially less of the released ²¹⁹Rn will reach air.

As can be seen in Figure 1.3, a radon isotope (218Rn) is also formed from 218At during the decay of 222Rn to stable lead. However, this decay is of little significance because only about 0.02% of the 218Po decays to 218At and 0.1% of this 218At decays to 218Rn, so that only about 2x10-5% of all 222Rn atoms present yields a 218Rn atom. Since at the same time the half-life of this radon isotope is 35 ms, it is of little significance in comparison with the other isotopes.

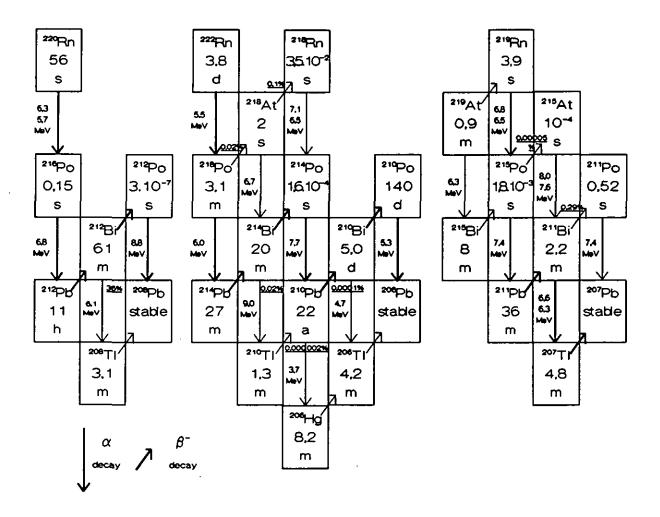


Figure 1.3. The decay of the radon isotopes to stable isotopes of lead; the alpha and beta decay, the energies of the alpha particles (MeV) and the half-lives are shown; less important transitions are indicated with a thin arrow along with the percentage of the nuclide concerned.

If only half-lives are taken into consideration, then nearly all the short-lived daughters of ²²²Rn and ²²⁰Rn in air are in radioactive equilibrium with their respective parent nuclides. Because these daughter products are chemically active, they readily attach to aerosol particles and the airborne concentrations of the unattached daughters decrease through plateout, so that the equilibrium becomes disturbed. Consequently, for a given activity concentration of radon in air, the air concentrations of its daughters can vary considerably, depending on such factors as aerosol concentration and ventilation rate.

Since the radiation dose received by humans from radon arises mainly from these daughter products, conversion to an equilibrium equivalent concentration (EEC) is desirable. This EEC is defined as that activity concentration of ²²²Rn or ²²⁰Rn in equilibrium with the short-lived daughters which corresponds to the same potential alpha energy of the daughters as the non-equilibrium mixture. It is the potential alpha energy which produces the ultimate radiation dose. The potential alpha energy of a daughter nuclide in the decay chain of ²²⁰Rn or ²²²Rn is the total alpha energy emitted during the decay of this nuclide to ²⁰⁸Pb and ²¹⁰Pb, respectively. The equilibrium factor F is defined as the ratio of the equilibrium equivalent radon concentration to the actual radon concentration:

$$F_{222Rn} = EEC_{222Rn} \left[^{222}Rn\right]^{-1}$$
 (1.11)

$$F_{220Rn} = EEC_{220Rn} [^{220}Rn]^{-1}$$
 (1.12)

Conversion of the radon concentrations to equilibrium equivalent concentrations therefore amounts to the calculation/estimation of the equilibrium factor F. This has been worked out for the daughters of ²²²Rn and ²²⁰Rn in Table 1.3. When ²²²Rn or ²²⁰Rn and its decay products are in radioactive equilibrium, all have the same activity concentration. The equilibrium equivalent concentration can then also be expressed in the potential alpha energy concentration (PAEC) per unit of activity (Table 1.3), expressed in J m⁻³ Bq⁻¹:

$$PAEC_{222Rn} \approx 5.54 \times 10^{-9} \times EEC_{222Rn}$$
 (1.13)
 $PAEC_{220Rn} \approx 7.57 \times 10^{-8} \times EEC_{220Rn}$ (1.14)

$$PAEC_{220Rn} \approx 7.57 \times 10^8 \times EEC_{220Rn}$$
 (1.14)

The relative significance of the daughter nuclides in the EEC is shown in the last column of Table 1.3. When the potential alpha energy of each daughter is used as a weighting factor, the equilibrium equivalent concentration can be given in terms of the concentrations of the most important daughters:

$$EEC_{222Rn} \approx 0.105 \text{ x } [^{218}Po] + 0.514 \text{ x } [^{214}Pb] + 0.381 \text{ x } [^{214}Bi]$$
 (1.15)

$$EEC_{220Rn} \approx 0.913 \text{ x } [^{212}Pb] + 0.087 \text{ x } [^{212}Bi]$$
 (1.16)

1.2.3 Emanation and exhalation of radon

Before radon is exhaled from the soil or building material, a process called emanation takes place. Emanation is defined as the release of the radon atoms from the solid particles of the source material. Once radon has entered the pores of the soil or building material, it is further transported by convection and diffusion. Exhalation refers to the release of radon from the surface of the soil or building material to air.

Table 1.3. Potential alpha energy of short-lived daughter nuclides of 202Rn and 202Rn (see Figure 1.3) per atom and per unit of activity (Jacobi et al., 1987; Radiological Health Handbook, 1970)

adionuclide	Relative proportion	Potential alpha energy (ϵ_p)				
		pe: MeV	r atom 10 ⁻¹² J	per unit of MeV Bq ⁻¹ 10	activity D ⁻¹⁰ J Bq ⁻¹	
	_	V ·	-			
² Rn daughte	rs:					
²¹⁸ Po	100	13.7	2.19	3620	5.8	
²¹⁸ At	0.0185	7.70	1.23	22	3.5x10 ⁻²	
²¹⁸ Rn	1.85×10 ⁻⁵	14.8	2.37	7.5x10		
²¹⁴ Pb	99.9815	7.69	1.23	17,800	28.5	
²¹⁴ Bi	100	7.69	1.23	13,200	21.1	
^{· 214} Po	99.979	7.69	1.23	1.8x10	-3 2.9x10 ⁻⁶	
²¹⁰ Tl	0.021	0	0	0	0	
otal for 222	Rn:			34,600	55.4	
⁰ Rn daughte	rs:					
²¹⁶ Po	100	14.6	2.34	3.16	5.1x10 ⁻³	
²¹² Pb	100	7.8	1.25	431,000	691	
²¹² Bi	100	7.8	1.25	40.900	65.5	
²¹² Po	64	8.79	1.41	3.9x1	0 ⁻⁶ 6.2×10 ⁻⁹	
²⁰⁸ Tl	36	0	0	0	0	
****	-					
otal for ²²⁰ 1	Rn:			472,000	757	

Emanation

Radon is produced in nature by the decay of radium, the parent nuclide. Like earlier members of the uranium and thorium decay series, radium is trapped in solid particles of the material under consideration (soil or building material). Diffusion of radon in these particles is very slow (L = 10⁻¹³-10⁻³² m according to Nazaroff and Nero, 1988). Therefore, there must be another process permitting the escape of radon atoms from the solid particles. This process is called "recoil". During the decay of ²²⁴Ra to ²²⁰Rn and ²²⁶Ra to ²²²Rn, alpha particles are emitted possessing energies of 5.7 and 4.8 MeV, respectively. These particles have a recoil effect on the radon atoms formed, which can cause them to migrate over a distance of 20 to 70 nm in common minerals and thereby eject them into the interstitial pores of the material (see Figure 1.4). This so-called recoil distance in air is 63 μ m for ²²²Rn and 83 μ m for ²²⁰Rn (Nazaroff and Nero, 1988), so that the released atom can readily penetrate a nearby soil particle again. It is possible that the atom will end up in the pores between the particles via the "pocket" created in the soil particle by diffusive transport (indirect recoil). If this interstitial space contains water, the radon atoms will be slowed down in this water before reaching another particle (this is because the recoil range in water is only 0.1 μ m), and can reach air-filled pores by diffusion. For a better understanding of the measured emanation rates, the description of the recoil process must be expanded. Two hypotheses have been advanced for which some support has been obtained (Nazaroff and Nero, 1988):

- . radium-226 and also ²²⁴Ra could have been deposited primarily in the surface layers of the soil particles;
- as a result of chemical corrosion and radiation damage (due to decay of the nuclides of the primordial decay series), the emanation range (the area in the soil

particles from which the recoil effect can occur) could be considerably increased due to the greater effective surface area of the particles.

The emanation coefficient η , the fraction of radon generated in soil which leaves the solid grains and enters the pore volume of the soil, lies for different soils within the range of 0.01 to 0.8 (UNSCEAR, 1988), depending on such factors as moisture content, corrosion and particle size.

A similar consideration applies to building materials. However, owing to the relatively dry nature of building materials as compared with soil, the emanation coefficients are usually slightly lower. Building materials in which fly ash is the main constituent have extremely low emanation coefficients ($\eta \sim 0.002-0.02$). This is because fly ash has a vitrified structure, from which escape of radon is virtually impossible.

Exhalation

The exhalation rate (E) from a soil or building material is determined by emanation and subsequent transport (diffusion and convection) of radon through the soil or building material. The exhalation rate is determined primarily by:

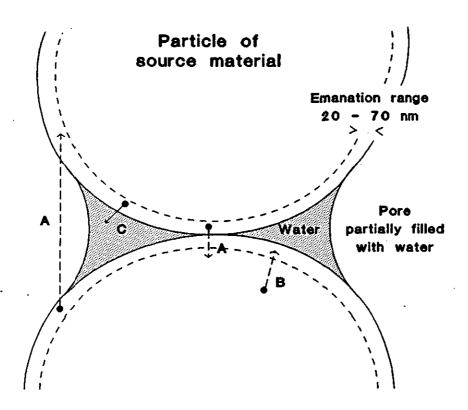


Figure 1.4. Emanation of radon from the source material; this figure shows (A) recoil from one particle into another, (B) recoil within one particle, and (C) recoil into interstitial water.

- the radium concentration in soil or building material
 temperature
 atmospheric pressure differences
 permeability
 moisture content (groundwater)
 porosity
 important for diffusion
- porosityemanation coefficient

The exhalation rate from soil due to diffusion is expressed by the formula:

$$E_{soil} = \lambda_{Rn} \eta C_{soil,Ra} \rho_{soil} L_{Rn}$$
 (1.17)

where $C_{soil,Ra}$ is the activity concentration of radium in soil (Bq kg⁻¹), ρ_{soil} (kg m⁻³) is the soil density, and L_{Rn} is the diffusion length of the radon isotope in soil (m) (see equation (1.2)). The range for the area exhalation rate from soils is reported by UNSCEAR (1988) to be 0.0002-0.07 Bq m⁻² s⁻¹ for ²²²Rn and 0.015-5.3 Bq m⁻² s⁻¹ for ²²⁰Rn.

Similarly, the exhalation rate from building materials due to diffusion can be calculated using the equation (Nazaroff and Nero, 1988; UNSCEAR, 1988):

$$E_{\text{build}} = \lambda_{\text{Rn}} \quad \eta \quad C_{\text{build},\text{Ra}} \quad \rho_{\text{build}} \quad L_{\text{Rn}} \quad \tanh[0.5\text{d} / L_{\text{Rn}}] \tag{1.18}$$

where $C_{build,Ra}$ is the activity concentration of radium in the building material (Bq kg⁻¹), ρ_{build} (kg m⁻³) is the density of the building material, and d is the thickness of the building material (m). Ranges of 0.00005-0.005 Bq m⁻² s⁻¹ for ²²⁰Rn and 0.01-0.1 Bq m⁻² s⁻¹ for ²²⁰Rn have been reported by Folkerts et al. (1984) for the area exhalation rate from building materials.

It can be concluded that in a moist soil (not dry, not saturated, as are many soils in the Netherlands), the emanation coefficient is maximal. Consequently, both diffusive (see Figure 1.4) and convective transport are important, so that radon exhalation from soil is maximal (Cothern and Smith, 1987). Depending on the grain size of the soil, diffusion or convection is the principal transport process. Building materials have slightly lower emanation coefficients than soils due to their low moisture content and relatively high density, and diffusion is an important transport process for ²²²Rn and ²²⁰Rn. Building materials based on fly ash have extremely low emanation coefficients, so that external irradiation due to radionuclides in these materials becomes the dominant exposure route. However, if pressure differences occur across building materials in buildings, convective transport of radon could remain a possibility.

1.3 STANDARDS AND GUIDELINES

The policy area of standards and guidelines for exposure to radon and radon daughters is showing much activity in many countries. However, not only are there uncertainties with respect to the dose-effect relationships for both ²²²Rn and ²²⁰Rn (see Chapter 6), but prediction of the concentrations of these two isotopes in the indoor atmosphere is as yet not possible for a specific building. For the time being therefore, regulation on an international level is taking place through the formulation of guidelines for maximum concentrations in existing and future housing. Since ²²²Rn and ²²⁰Rn are naturally occurring radioactive gases and are not easily controllable, if at all, in the outdoor environment (soil), policy at an international level concentrates on the possible control of the radon

concentrations in indoor environments (through entry from soil, building materials and any other indoor sources of radon). Radiation policy in the Netherlands is also aimed at the reduction of emissions from non-nuclear industries and functional applications (e.g., natural gas, drinking water) (see 1.3.2.). In several countries (though not in the Netherlands) there are specific standards for occupational exposure of miners to radon as well.

1.3.1 Units

The guideline for radon exposure is usually expressed as a time-integrated activity concentration or potential alpha energy concentration, expressed as the working-level month (WLM). One WLM corresponds to an exposure to one working level (WL) for one working month (170 h). One WL is the amount of potential alpha energy ϵ_p present in one cubic metre of air resulting in the emission of 2.08×10^{-5} J (arising historically from the potential alpha energy of 100 pCi ²²²Rn per litre (equivalent to 3700 Bq m⁻³ of ²²²Rn).

1 WL equals 3700 Bq
$$m^{-3}$$
 (EEC_{222Rn}) (1.19)

1 WL equals 270 Bq
$$m^{-3}$$
 (EEC_{220Rp}) (1.20)

and from the definition of one WLM it follows that

The guideline is sometimes also expressed as an annual effective dose equivalent (mSv a⁻¹). For the radon concentrations derived from this, it is usually assumed that an exposure to a concentration of 1 Bq m⁻³ (EEC_{222Rn}) results in an effective dose equivalent of 10⁻⁴ Sv a⁻¹. Residence times, breathing rates and lung dosimetry have already been taken into account in this implicit conversion factor (see Chapters 6 and 7 for dose-effect relationships).

1.3.2 Guidelines and standards for radon in the outdoor environment

There are no separate guidelines for radon in outdoor air. However, radon released by non-nuclear installations falls under the risk limits given in "Radiation protection and risk management" (VROM, 1991). Accordingly, mortality risks due to any radon and other radioactive emissions from a non-nuclear installation should not exceed a limit of 10⁻⁶ per year. The radioactivity released by various sources, such as gas-fired power stations, is mainly ²²²Rn, so that here only ²²²Rn has to meet the limit (²²⁰Rn will usually decay during transport through long gas pipelines). The maximum permissible risk level from cumulative sources is 10⁻⁵ per year. In the long run (the target year is 2000), however, a risk reduction to 10⁻⁸ per year per source (the negligibility level) is proposed for all sources from the categories "Functional applications" of radiation and "Non-nuclear industry" (VROM, 1991).

1.3.3 Guidelines and standards for radon in the indoor environment

Table 1.4 presents a few guidelines and standards which are in force in various countries. Sometimes there are different guidelines and/or standards in a multi-stage guidance system (collection of guidelines and standards):

- at ²²²Rn concentrations below a specified level (the basic level in Table 1.4), no remedial action is considered;

- at slightly higher ²²²Rn concentrations, simple remedial measures to bring these concentrations to below the basic level are examined;
- at ²²²Rn concentrations exceeding an upper limit (so-called action level), remedial actions should be seriously considered (sometimes in stages), depending on the seriousness of the situation and available control techniques.

This multi-stage guidance system generally applies to existing houses and often only for ²²²Rn. For future housing, lower guidelines are usually given as a basic level.

A multi-stage guidance system does not yet exist in the Netherlands. The concentrations measured in Dutch homes usually lie below the radon guideline given by, for example, the EC for future constructions (see Table 1.4). However, at present a standstill approach is being used in the Netherlands for building materials: "...in new dwellings (or those being replaced), no situations from the point of view of radiation protection are permitted than those which are worse than currently found in representative, postwar dwellings" (VROM, 1991). To support this standstill principle, research has been conducted into possible reference levels for building materials as part of the RENA (Controllable forms of natural background radiation) research programme, with respect to both radon exhalation and external radiation arising from radionuclides contained in building materials (e.g. van Heijningen and Ackers, 1990). These reference levels should eventually lead to product standards. After the publication of the Integrated Criteria Document Radon, a reduction objective for the long term will be formulated in 1991 aimed, for example, at improved construction practices for new buildings. Also, a standpoint will be defined as to the necessity and possibility of developing remediation criteria for the current housing stock (VROM, 1991).

Guidelines (basic and action levels) for the concentrations of ²²²Rn [Bq m⁻³ (EEC)] in the indoor environment as proposed by various organizations, or adopted (more-or-less stringent standards) in several countries (the original guidelines, if differing from those expressed in EEC, are given in parentheses)

Organization	Country	Existing homes Basic level Action level	Future homes	Comments	Reference
RPB	Canada	≈ 370 (0.1 WL)	considered unnecessary	Proposed in 1985	O'Riordan et al., 1987
RPC	Germany	≈ 200 (20mSv a ⁻¹)	qualitative approach	Proposed in 1986 if simple solution is possible	O'Riordan et al., 1987
EC Comm.	European Community	≈ 200 (20mSv a ⁻¹)	≈ 100 (10mSv a ⁻¹)	Recommendations as from 1990	EC, 1990
СМВ	Finland	≈ 360 (2µJ m ⁻³)	≈ 90 (0.5µJ m ⁻³)	Limit (standard) adopted in 1986	O'Riordan et al., 1987
wнo	international	100 400	100	Remedial action to be performed before added exposure exceeds 2000 Bq m ⁻³ a	WHO, 1987

IRCP	international homes workers 1: workers 2:		200 DAC:1500 DAC: 330 DAC:150,000 DAC:250,000	EEC (Rn) *) EEC (Tn) *) EEC (Rn) EEC (Tn)	Higher level for more difficult remedial actions Exposure for 2000 h a ⁻¹ in mines No daughters due to removal by exhaust or electrostatic deposition	ICRP, 1984 ICRP, 1986
RPA	Nordic countries		100	100	Proposed in 1986	O'Riordan et al., 1987
NRPB	United Kingdom		≈ 200 (20mSv a ⁻¹)	≈ 50 (5mSv a ⁻¹)	Proposed in 1987; the urgency of action depends on how high the concentration is adopted in 1987 (standard)	O'Riordan et al., 1987 McLaughlin, 1988
NCRP	United States		≈ 150 (2 WLM a ⁻¹) ≈ 185 (0.05 WL)		Proposed in 1984 for general application for 80% occupancy time	NCRP, 1984; 1987 NCRP, 1984
ЕРА			≈ 75 (0.02 WL) 37 - 185 (0.01-0.05 WL)		Adopted in 1986 (standard for certain areas); the urgency of action depends on how high the concentration is "Contaminated areas" (e.g. phosphate landfill).	O'Riordan et al., 1987 Nazaroff and Nero, 1988
NBHW	Sweden	200	400	70	Limit (standard) adopted in . 1980.	O'Riordan et al., 1987
NIRP		100 (re:	100 400 novation: 200)	100 70	Proposed in 1984 Below basic level if remedial action is fairly simple	O'Riordan et al., 1987 Swedjemark and Makitalo, 1990

^{*)} DAC = Derived Air Concentration: concentration which leads to the dose limit; EEC(Rn) = EEC_{222Rn}; EEC(Tn) = EEC_{220Rn}

For radon sources other than soil and building materials (so-called functional applications and non-nuclear industry), risk limits have been proposed. The proposed maximum permissible individual mortality risk due to natural gas and drinking water separately is 10^{-6} per year (see also 1.3.2).

Other limits apply to employees (including miners), namely, 2 mSv a⁻¹ (corresponding to a risk of $5x10^{-5}$ per year) for non-radiological workers and 20 mSv a⁻¹ (corresponding to a risk of $5x10^{-4}$ per year) for radiological workers (employees for whom a realistic dose estimate is an annual dose equivalent of more than 2 mSv). Incidentally, the principle applied here is the As Low as Reasonably Achievable (ALARA) principle.

2 SOURCES AND EMISSIONS

A radon source is a material from which radon is exhaled (e.g. soil, building material), or a location where radon emission occurs (e.g. non-nuclear industry, gas geyser, tap water). Radon-222 and ²²⁰Rn are decay products of two isotopes of radium, which are both present in the earth's crust. It is therefore not surprising that the soil is the most important source of radon. Other radon sources, such as groundwater and building materials, are usually sources because they have been in direct contact with the soil, or are made out of natural materials. This chapter describes the various sources and their emissions.

2.1 EXHALATION FROM THE SOIL

Exhalation of radon from the soil is affected by several factors (see 1.2), which are listed in Table 2.1. External factors, such as atmospheric pressure differences and temperature, can be important for convective transport through the soil. Consequently, the radon exhalation rate is dependent on the soil type and various meteorological conditions, such as rainfall, humidity, evaporation, temperature, pressure changes, etc. The resulting area exhalation rate usually lies in the range of 0.0002 to 0.07 Bq m⁻² s⁻¹ for ²²²Rn and 0.015 to 5.3 Bq m⁻² s⁻¹ for ²²⁰Rn (based on UNSCEAR, 1988). Assuming an average exhalation rate of 0.016 Bq m⁻² s⁻¹ for ²²²Rn and of 1-2 Bq m⁻² s⁻¹ for ²²⁰Rn (based on UNSCEAR, 1988), which, in view of the measured values, seems a reasonable assumption for the Netherlands (see Ackers, 1985 and Chapter 4), and using a land area of 38,000 km² (CBS, 1990), the resulting annual exhalation rate from the soil is found to be 1.9x10¹⁶ Bq of ²²²Rn and 1.4x10¹⁸ of ²²⁰Rn. Since 8% of the Netherlands is covered (infrastructure and buildings) and housing constitutes only part of this, approximately 1% of this radon will enter dwellings directly through the foundation or indirectly with outdoor air.

Table 2.1. Factors important in determining radon exhalation from soil; a
typical value and the characteristic range are given (see 1.2)

Factor	Typical value	Characteristic range	Unit
Radium concentration **)	25	4 -125	Bq kg ^{.1}
Emanation coefficient	0.2	0.01 - 0.8	2 1
Diffusion coefficient	10⁴	5x10 ⁻⁶ - 2x10 ⁻¹⁰	m²s-1
permeability (sandy clay)	~ 10-14	10 ⁻⁸ - 10 ⁻¹⁶	m ²
Porosity	0.5	0.45 - 0.55	
Percentage of water-filled pores	d 50 *)	0 -100	*

^{*)} highly dependent on the permeability and the distance to the water table

2.2 EXHALATION FROM SEA AND SURFACE WATERS

Because of the relatively low radium activity concentration in sea and surface waters (see Table 2.2 for 226 Ra) compared to that in sediment or in soils (25 kBq m⁻³), and because of the much lower diffusion rate in water as compared to that in soil (D $\approx 10^{-9}$ m² s⁻¹ and $\approx 10^{-6}$ m² s⁻¹, respectively), the radon exhalation rate is also much lower. According to the NCRP (1984a), the exhalation rate from the oceans is 10^{-4} Bq m⁻² s⁻¹, or a factor of 160

^{**)} Iyengar, 1990

lower than that from the soil. This will also be a reasonable estimate for the other types of deep stratified water bodies. In general, the concentrations of ²²⁴Ra (the parent of ²²⁰Rn) in ocean surface water are slightly lower (0.2-0.7 Bq m³) than those of ²²⁶Ra, whereas in coastal waters, especially in shallow estuaries, higher concentrations have been observed (3-15 Bq m⁻³) (Okubo, 1990). Because of the lower concentrations of ²²⁴Ra as compared to those of ²²⁶Ra, the exhalation rate of ²²⁰Rn from the oceans will be no more than a factor of 10-15 higher than that of 222Rn. The situation is somewhat different for vertically wellmixed rivers and non-stratified lakes. The radon exhaled from the sediment is mixed in the water column above it and can escape from the surface to the atmosphere. Diffusion does not play an important role in this case. Consequently, the radon concentrations in these types of water bodies be higher than would be expected from the radium concentration. The literature gives activity concentrations of about 300 Bq m⁻³ for ²²²Rn for such situations in the United States (Nazaroff et al., 1987). Because of the short halflife of ²²⁰Rn, this mixing will not greatly influence the exhalation rate of this isotope. Owing to the relatively low radon concentration in marine air, it may be expected that the annual average radon concentrations in air over maritime countries, such as the Netherlands, are lower than over areas farther away from the coast (see Chapter 3).

Table 2.2. Radium-226 concentration in some sea and surface waters

Type of water	²²⁶ Ra concentration Bq m ⁻³	Reference
Atlantic Ocean (surface)	1.3	Okubo, 1990
Continental surface waters	0.08 - 300	Beneš, 1990
Estuaries	≈ 3	Moore, 1990
Continental rivers	2.6	Iyengar, 1990
Rhine in Germany	3.3 - 7.4	Iyengar, 1990
Lakes (Europe)	0.5 - 7.4	Iyengar, 1990

2.3 FOSSIL FUELS AND THEIR COMBUSTION

By burning coal, oil and natural gas, nearly all the radon contained in it is released. However, enhancement of the natural activity from the raw material to the ash produced (coal) may have occurred. This ash constitutes a permanent source of radon. The radon emissions from burning the principal conventional fuels in the Netherlands are considered in more detail below.

2.3.1 Coal

Coal consumption in the Netherlands in 1988 was approximately 1.3×10^{10} kg (Blaauboer et al., 1991), producing about 1.5×10^9 kg of ash (bottom ash, fly ash and dust). During combustion the ²²²Rn initially present in the coal, about 3.2×10^{11} Bq, is released. Given the activity concentrations of the parent nuclides, the amount of ²²⁰Rn discharged to the atmosphere will be similar. However, coal ash also contains 3.2×10^{11} Bq of ²²⁶Ra and ²²⁴Ra. A substantial proportion of this ash is used in the manufacture of building materials, and as road fill. However, because fly ash particles are in a vitrified form, not much of the ²²²Rn produced by the decay of ²²⁶Ra in the source material can be released (about 1%). This will probably be even less for ²²⁰Rn due to its short half-life. Nevertheless, higher exposure rates can be measured near such building materials and also above asphalt mixed with fly ash as a result of photon emission. Elevated ²²²Rn concentrations have

never been found in the vicinity of coal-fired power stations (Corbett, 1983).

2.3.2 Petroleum

The ²²²Rn and ²²⁰Rn contained in petroleum has usually already been released from this material before it arrives in the Netherlands. Besides, the activity of the primordial radionuclides in petroleum is much lower than in coal, and these nuclides are dispersed during combustion (no fly ash!). The use of oil (in the form of fuel oil) in electric power stations in the Netherlands has declined sharply over the past few years to a few per cent of that of coal (CBS, 1990).

2.3.3 Natural gas

All the ²²²Rn and ²²⁰Rn contained in natural gas escapes during its combustion. The natural gas used in the Netherlands comes from gas fields in the North Sea and on the mainland (e.g. Slochteren in the Netherlands). Most of the ²²⁰Rn in natural gas will have decayed during transit from producers to consumers. External irradiation can occur at distribution centres in the gas pipelines because of buildup of radon daughters. Neglecting the transit time for ²²²Rn, and taking an average ²²²Rn concentration in natural gas of 100 Bq m⁻³ and a consumption of 7.9x10⁹ m³ a⁻¹ by the gas-fired power stations (Blaauboer et al., 1991), the atmospheric emission of ²²²Rn in 1988 will have been nearly 8x10¹¹ Bq. The use of natural gas by other consumers (mainly households and industry) amounted to about 3x10¹⁰ m³ in 1988 (CBS, 1990), producing an additional ²²²Rn emission of 3x10¹² Bq in that year. The latter emission was chiefly directed to the outdoor atmosphere, but in enclosed rooms with unvented gas water heaters (households), the ²²²Rn emission directly entered the indoor atmosphere. These emissions fall under the risk limits for "Non-nuclear industry" proposed in the policy paper "Radiation protection and risk management" (VROM, 1991).

2.4 MINERALS

The total 226 Ra throughput in the processing industry is 4×10^{12} to 5×10^{12} Bq a⁻¹, the principal sectors being phosphoric acid, fertilizers, and iron and steel (Blaauboer et al., 1991). The 222 Rn formed from the decay of this 226 Ra is about 3×10^{14} Bq a⁻¹. It should be noted that a substantial proportion of the radium activity is discharged into surface water, so a large fraction of the 222 Rn formed will not enter the atmosphere. Insofar that ore tailings are dumped on land, emanation and diffusive transport must also be taken into consideration. Since 226 Ra ($t_{1/2} \approx 1600$ years) will produce 222 Rn for many years, allowance must be made for buildup of radium and thorium. In the vicinity of disposal sites of slag from phosphate-ore processing, which contains 1500 Bq kg⁻¹ of 226 Ra, elevated 222 Rn concentrations can therefore be expected (no data are available for 220 Rn). This is true to an even greater extent for uranium tailings. Uranium is not mined in the Netherlands, but uranium enrichment is carried out at Urenco (Almelo). However, the depleted uranium is always returned, along with the enriched uranium, to Urenco's customers, mainly located abroad.

Another source of radon are phosphate fertilizers. The amount of phosphate fertilizer applied annually per hectare of agricultural land is on average 50 kg, resulting in a total addition of 10⁸ kg a⁻¹ to the Dutch agricultural soils (Langeweg, 1988). A typical value for the ²²⁶Ra concentration in these fertilizers is 1000 Bq kg⁻¹ (UNSCEAR, 1988). Consequently, the annual addition to the soil activity is 5 Bq m⁻² ²²⁶Ra, or 10¹¹ Bq on a national scale. When the soil is ploughed to a depth of 30 cm, the ²²⁶Ra concentration in

this layer increases annually by 5 Bq per 12,000 Bq of natural ²²⁶Ra present, or has increased by about 2.5% since the use of phosphate fertilizers began in 1920 (Langeweg, 1988). This generates about $4x10^{14}$ Bq of ²²²Rn per year. Only a fraction of this will be released into the atmosphere, so that fertilizer application results in a very small increase of the background level.

Fairly large amounts of radon can also be released during ore or coal mining (especially when the ores are rich in uranium/thorium). Here the problem usually is that radon concentrations remain high in the air in the mine tunnels and so constitute a source of exposure to the miners. Mining was discontinued in the Netherlands itself in 1963, but Dutch miners still work in Belgian and German coal mines.

2.5 BUILDING MATERIALS

Because the constituents of building materials (such as brick and concrete) are derived directly or indirectly from the earth, they too usually have radium concentrations equal to or sometimes even greater than those in the ground. An example of the latter is phosphogypsum, a by-product of the phosphate ore-processing industry, which has concentrations exceeding those in the soil by a factor of 30. The radon exhalation rate from building materials is determined by several factors (see 1.2.3), so that the total annual emission of ²²²Rn and ²²⁰Rn from construction materials in the Netherlands is difficult to assess. This is partly due to the fact that decorative coatings applied to these building materials, such as wallpaper, panelling and paint, tend to reduce the radon exhalation rate (Van Dijk and De Jong, 1989). However, a rough estimate of the total emission can be made on the basis of a surface area (inside + outside) of 750 m² for a reference house (UNSCEAR, 1988: adapted from Table 14, p.103), a total of five million dwellings and a typical exhalation rate from building materials of about 5x10⁻⁴ Bq m⁻² s⁻¹ of ²²²Rn (see Table 5.3). The total annual emission of ²²²Rn from building materials used in the construction of dwellings would then be:

$$5x10^6 \times 750 \text{ m}^2 \times 5x10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1} \times 3.15x10^7 \text{ s a}^{-1} \approx 5.9x10^{13} \text{ Bq a}^{-1}$$
.

The exhalation rate of ²²⁰Rn is a factor of 77 higher (see 1.2.3). Approximately 60% of the emission enters the indoor atmosphere (UNSCEAR, 1988). Taking into account building materials which have been incorporated into public buildings, office blocks, factories, etc., the emission might even be a factor of 2 higher. The total amount exhaled from building materials is then 1 to 2% of that from soil.

2.6 GROUNDWATER

Groundwater may be a significant source of indoor airborne radon (mainly ²²²Rn, because of its relatively long half-life and the transit time from the drinking-water-supply plants to homes). Since groundwater is in direct contact with radium-bearing soil and the high concentrations of radon gas in it, the radon concentrations in groundwater are as a rule much higher than in surface water. Radon-222 concentrations in groundwater have been observed to range from 1 Bq m⁻³ to 100,000 Bq m⁻³ worldwide (Nazaroff and Nero, 1988; Nazaroff et al., 1987; Prichard, 1987). A population-weighted average radon concentration of about 5200 Bq m⁻³ has been estimated in the United States (Nazaroff et al., 1987). When groundwater is used for the production of drinking water, but especially when water is drawn from private wells, the tap water can have high ²²²Rn concentrations.

Part of the ²²²Rn is transferred to indoor air (about 55% according to Nazaroff et al., 1987), for example, as a result of aeration or rise of temperature (e.g. during a hot shower). The ²²²Rn concentrations in Dutch drinking-water supplies have not been extensively investigated. Using an average concentration of 4800 Bq m⁻³ in groundwater (Mattern, 1973), an average water use of 0.2 m³ per day per person (CBS, 1990), a user group comprising 70% of the Dutch population (CBS, 1990), or about 10 million persons, and a ²²²Rn transfer efficiency from drinking water to indoor air of an average of 55%, the total emission of ²²²Rn into the indoor environment due to drinking water can be calculated to be:

4800 Bq m⁻³ x 0.2 m³ d⁻¹ x 10⁷ x 365 d a⁻¹ x 0.55
$$\approx$$
 1.9 x 10¹² Bq a⁻¹

In areas with higher groundwater concentrations, but especially in homes using private wells, the concentrations in drinking water can be higher. When using surface water supplies (which serve about 30% of the Dutch population), the expected ²²²Rn concentrations will be much lower (see 2.2).

Radon emission into air also arises from evaporation of groundwater. The principal process by which this evaporation occurs is uptake by, and transpiration from, vegetation (see 2.7.3).

2.7 OTHER SOURCES

Various other sources of radon, which as a rule make a much smaller contribution to radon in air than the ones mentioned above, are briefly discussed below.

2.7.1 Volcanism

Volcanoes and lavas are local sources of airborne radon. Lava, however, is newly formed rock which has a very low emanation coefficient, because radium is probably uniformly distributed in the source material and the mineral grains have a relatively small emanation range (see 1.2.3). During an eruption, ²²²Rn and ²²⁰Rn are released in addition to various other gases. Volcanic activity does not occur in the Netherlands, so that exposure results only from transcontinental movement of air masses.

2.7.2 Exhalation by humans

A minor, but unexpected source of radon comes from humans themselves. The annual dietary intake of ²²⁶Ra by adults is 16 Bq (Blaauboer et al., 1991), resulting in a body burden of about 1 Bq (UNSCEAR, 1988). The decay of this ²²⁶Ra yields about 70 Bq of ²²²Rn per year. The entire Dutch population produces annually around 10⁹ Bq of ²²²Rn. The dietary intake of ²²⁸Ra is similar to that of ²²⁶Ra, so that the production of ²²⁰Rn is 5.8x10¹² Bq a⁻¹. However, most of this ²²⁰Rn will decay in the body.

2.7.3 Exhalation by plants and animals

As in the case of humans, other living organisms also take up radium, but the magnitude of this intake is not known. Taking the biomass of the livestock in the Netherlands (CBS, 1990) and assuming a radium concentration which, in view of the concentration factors of radium in vegetation and meat (IAEA, 1982) is comparable to the concentration in humans, the amount of radon exhaled by this livestock will be no more than a few times that exhaled by humans.

Evaporation of groundwater occurs via water uptake by, and transpiration from, plants.

The amount thus evaporated in the Netherlands is about 500 mm a-1 (Kuipers, 1981), or:

$$38,000 \times 10^6 \text{ m}^2 \times 0.5 \text{ m}^3 \text{ a}^{-1} = 1.9 \times 10^{10} \text{ m}^3 \text{ a}^{-1}$$

At a ²²²Rn concentration in groundwater of on average 4800 Bq m⁻³, the resulting ²²²Rn emission by vegetation is about 9x10¹³ Bq a⁻¹. Radon-222 emissions have been measured from maize fields to air which were a factor of 3 higher than those from soil (Pearson, 1967).

2.8 TOTAL ²²⁰Rn AND ²²²Rn EMISSIONS IN THE NETHERLANDS

The calculated total emissions from the preceding sections are summarized in Table 2.3. It is seen that the emitted activity of ²²⁰Rn is much greater than that of ²²²Rn, but because of the often slow diffusion through building materials and the relatively short half-life of ²²⁰Rn, only a fraction of it will be released into indoor air. Because of its short half-life, the concentration of ²²⁰Rn in air 1 m above ground level will also already be lower than that of ²²²Rn (see 3.1.1). Soil is the principal source of ²²²Rn in outdoor air, its relative contribution being about 95%. The summary shows that only a few of the sources considered here result in a direct radon emission into the indoor environment: soil, building materials, natural gas, drinking water, and humans themselves. The total direct emission of ²²²Rn into indoor air contributes only about 2% to the total ²²²Rn emission. However, since radon accumulates in indoor spaces and most individuals spend a large amount of time indoors, these emissions are of greater importance for the radiation dose to humans than, for example, the uncontrollable exhalation from soil to the outdoor atmosphere. The principal sources of indoor 222Rn are the soil under and near the building (relative contribution about 70%) and building materials (about 30%). The other sources contribute on average about 1% to indoor 222Rn. In view of the above findings, the emphasis in this integrated criteria document is on the indoor environment and the sources which are important here. Relatively little attention is paid to radon in the outdoor environment.

Table 2.3. Emissions of 220Rn and 222Rn in the Netherlands

Source	²²⁰ Rn	²²² Rn		emission of ²²² Rn	
	Bq a-l	Bq a ⁻¹	into t	he indoor en Bq a ⁻¹	nvironment % (b)
Soil	1.4x10 ¹⁸	1.9x10 ¹⁶	≈ 1	≈ 2x10 ¹⁴	≈ 70
Volcanism	na *)	na		- *)	
Coal combustion	3.2x10 ¹¹	3.2x10 ¹¹		- ′	
Fly ash (1 year)	nk *)	1.1x10 ¹¹		_	
Natural gas	na	3.8x10 ¹²	< 40	$<1.5\times10^{12}$	< 0.5
Fuel oil	na	na		_	
Phosphate-ore tailings	nk	<3.0x10 ¹⁴		-	
disposed of in	nk	nk		-	
earlier years	nk	nk			
Phosphate fertilizers	nk	6.7×10^{12}		-	
used in earlier year		$<4.0x10^{14}$		-	
Building materials	9.2x10 ¹⁵	1.2x10 ¹⁴	60	$\approx 7.5 \times 10^{13}$	≈ 30
Groundwater					
domestic use	nk	1.9x10 ¹²	100	1.9x10 ¹²	≈ 0.6
evaporation	nk	9.0x10 ¹³		_	
Surface water					
domestic usé 1)	nk	5.9x10 ¹⁰	100	5.9x10 ¹⁰	≈ 0.02
Exhalation					
humans	<<5.8x10 ¹²	1.0x10°	80²)	8.0x10 ⁸	_
	<<1.7x10 ¹³	3.0x10 ⁹	•	-	
vegetation	nk	nk		nk	
Total	≈ 1.5×10 ¹⁸	2.0x10 ¹⁶	≈ 1.5	≈ 3x10 ¹⁴	100

^{*)} nk = not known; na = not accounted for; - negligible

2.9 POLICY ASPECTS OF EMISSIONS OF ²²⁰Rn AND ²²²Rn

In accordance with the policy paper "Radiation protection and risk management", the radon emissions can be divided into four source categories, namely, "Natural", "Functional applications", "Non-nuclear industry", and "Building construction and occupancy" (VROM, 1991). This classification is presented in Table 2.4 for the sources discussed earlier in this chapter. Insufficient information is available for ²²⁰Rn. Because of the shorter half-life of ²²⁰Rn, the various exposure pathways will probably be more easily controllable for this radon isotope than for ²²²Rn. The category "Natural" is not a subject of the setting of risk limits in radiation protection policy. The risk approach (a maximum of 10⁻⁶ per source) applies to the categories "Functional applications" and "Non-nuclear industry" (see 1.3.2 and 1.3.3). In view of the contributions from soil and building materials to the total radon emission (see Table 2.3), the category "Building construction and occupancy" is in this case the most important one. Besides an already existing standstill principle for building materials, a more precise position on reduction objectives (see 1.3.3) for radon concentrations in indoor air at a later stage will, for example, be formulated on the basis of this document.

¹⁾ a 222Rn concentration of 300 Bq m⁻³, and serving 30% of the population

²⁾ assuming an indoor occupancy factor of 0.8 (UNSCEAR, 1988) a) percentage of the source contribution to the total emission which enters the indoor environment

b) per cent contribution to the total emission into indoor air

Classification in terms of policy (VROM, 1991), relevant exposure pathways and an indication of the controllability of $^{222}\!\mathrm{Rn}$ emissions in the Netherlands Table 2.4.

Source category	Relevant exposure pathway	Controllability indication ')
Functional applications	· · · · · · · · · · · · · · · · · · ·	
Radioactive waste		•
- uranium tailings	exhalation → outdoor air	none 2)
Non-nuclear industry		
Coal combustion	emission → outdoor air	poor
Fly ash (1 year)		
- dumps, roads	exhalation → outdoor air	none
building materials	exhalation → outdoor/indoor air	moderate 2)
Natural gas		
- transport	external irradiation	moderate 2)
energy production	emission → outdoor air	poor
- domestic use	emission → indoor air	good
Fuel oil	emission → outdoor air	
- energy production	emission → outdoor air	poor
Groundwater - domestic use	emission → indoor air	poor
Surface water	emission - indoor all	poor
- domestic use	emission → indoor air	poor
Phosphate ore-processing	emission → indoor air	poor
industry		F
Wastes		
- from process	emission → outdoor air	poor
- earlier years	exhalation → outdoor air	moderate
Phosphate fertilizers	exhalation → outdoor air	moderate
 use in agriculture 	exhalation → outdoor air	none
earlier years	exhalation → outdoor air	none
Building construction and oc	CUDANCY	
Building materials	exhalation → indoor/outdoor air	mod./good
Soil	exhalation → indoor air	moderate
Natura 1		
Soil	exhalation → outdoor air	none
Volcanism	emission → outdoor air	none 3)
Groundwater	evaporation→ outdoor air	none
Surface water	exhalation → outdoor air	none
Exhalation by:		
- humans	exhalation → indoor/outdoor air	none
- animals	exhalation → (indoor)/outdoor as	r none
vegetation	exhalation → outdoor air	none

¹⁾ none

none : no possibilities
poor : only after closure of industry or other far-reaching measures
moderate: limited possibilities

good : good possibilities

2) exposure, especially of workers

3) Emissions do not occur in the Netherlands.

2.10 SUMMARY AND CONCLUSIONS

Various sources contribute to the emission of ²²²Rn and ²²⁰Rn. Apart from the soil itself, nearly all the sources discussed in this chapter are derived from the ground or have been in contact with it. Soil contributes by far the most (≈ 95%) to the total emissions (≈ $2x10^{16}$ Bq a⁻¹ of ²²²Rn and $\approx 1.5x10^{18}$ Bq a⁻¹ of ²²⁰Rn). Phosphate fertilizers applied to the Dutch soil over the years and ore tailings are the second largest source ($< 7x10^{14}$ Bq a⁻¹ of ²²²Rn; emission of ²²⁰Rn not estimated). The third largest source are the building materials ($\approx 1.2 \times 10^{14}$ Bq a⁻¹ of ²²²Rn and $\approx 9.2 \times 10^{15}$ Bq a⁻¹ of ²²⁰Rn). The remaining sources together produce about 10¹⁴ Bq a⁻¹ of ²²²Rn (production of ²²⁰Rn not estimated). Radon in indoor air ("Building construction and occupancy") makes the largest contribution to the total radiation dose to humans because of the accumulation indoors especially of ²²²Rn. The concentrations of ²²²Rn and ²²⁰Rn in indoor air are determined by the exhalation rate from soil and from building materials. A change in the former over time is not to be expected. There is a possibility of increasing emissions from the use of industrial by-products in the manufacture of building materials. Implementation of the standstill principle (see 1.3) and the development of product standards for building materials may affect this trend.

3 BEHAVIOUR AND DISPERSION

The first part of this chapter gives a general description of the behaviour of radon in the atmosphere, with attention being paid to the dispersion and deposition of its daughters. Next, the movement of radon in soil and its entry into buildings are discussed. Finally, a number of examples are given of organisms which accumulate ²¹⁰Pb and/or ²¹⁰Po derived from ²²²Rn in the atmosphere.

The data provided in this chapter are, in principle, universally applicable and for the most part do not specifically pertain to the Netherlands. For more information about the Dutch situation the reader is referred to Chapter 5.

3.1 DISPERSION IN THE ATMOSPHERE AND DEPOSITION OF RADON DAUGHTERS

3.1.1 Dispersion in the atmosphere

Chapter 2 has shown that the principal source of ²²²Rn and ²²⁰Rn (thoron) in air above land surfaces is the soil. Because of this the dispersion of radon, unlike that of many other substances, is characterized by an area as the source and not by one or more point sources. Local differences in the exhalation rate from soil lead to differences in concentrations at ground level, particularly when there is no wind. The transport and dispersion of radon in the atmosphere is determined by diffusion and convection, in addition to the predictable influence of radioactive decay. Convective radon transport is strongly influenced by meteorological factors, such as vertical temperature gradient, wind speed and direction, and vertical turbulent mixing. Since the decay products of radon attach to aerosols, their dispersal is also affected by dry and wet deposition. Because of the large difference between the half-lives of ²²²Rn (3.8 d) and ²²⁰Rn (56 s), the dispersion of ²²⁰Rn is confined to the source area. Radon-222 is one of the substances which are dispersed on a continental scale.

On the basis of model calculations, figure 3.1 gives an idea of the dependence of ²²²Rn and ²²⁰Rn concentrations on altitude, normalized to the exhalation rate from the ground, for different atmospheric stabilities (UNSCEAR, 1988). It shows that ²²²Rn, owing to its longer half-life, is dispersed to much higher altitudes than ²²⁰Rn. At 10-100 m height the ²²⁰Rn concentrations are insignificant compared with those at ground level. Radon-222 rises to an altitude of more than 10,000 m. For ²²²Rn, the results of model calculations are in reasonable agreement with those of the measurements. For ²²⁰Rn, this has been confirmed only for heights up to about 10 m (UNSCEAR, 1982). Actual data on ²²⁰Rn distribution with height over a few hundred metres to compare with the models are not available.

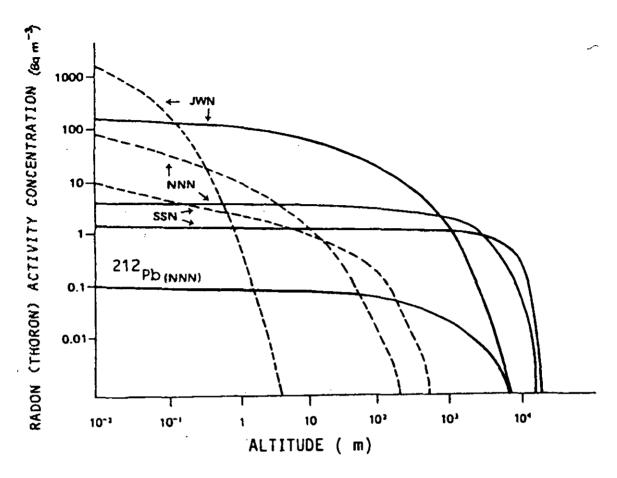


Figure 3.1. Vertical profiles of ²²²Rn (solid lines) and ²²⁰Rn (broken lines), based on exhalation rates of 0.02 Bq m⁻² s⁻¹ for ²²²Rn and 1.0 Bq m⁻² s⁻¹ for ²²⁰Rn, and different atmospheric stabilities (JWN = strong inversion; SSN = strong turbulence; NNN = normal turbulence)

It is not possible to make a general statement about the seasonal variations of the radon concentration at ground level. On the one hand, the exhalation rate of radon from the ground generally shows a seasonal variation, due to the fact that the upper soil layers contain more water and the water table is higher during the winter than the summer, resulting in decreased exhalation. On the other hand, vertical turbulent mixing is usually higher in summer than in winter, resulting in greater upward dispersion. In some areas of the Northern Hemisphere, such as New Jersey in the USA, the effect of the decreased exhalation rate outweighs that of decreased mixing and the concentration is lower in the winter than in summer. Different seasonal patterns have been observed at other locations. The atmospheric concentration at coastal sites is strongly influenced by the wind rose.

In addition to seasonal variations, the ²²²Rn and ²²⁰Rn concentrations also show a diurnal variation. The concentrations generally reach a maximum at night and a minimum at noon or in the afternoon. This has been demonstrated in such countries as the USA (see Figure 3.2), the former USSR, France, Hungary and India. (UNSCEAR, 1982; Cothern and Smith, 1987). The main causes are probably the lower wind speeds and the stronger inversion during the night which greatly reduces the convective transport to higher layers of the atmosphere. Heating of the ground by the sun also plays a part, creating an upward

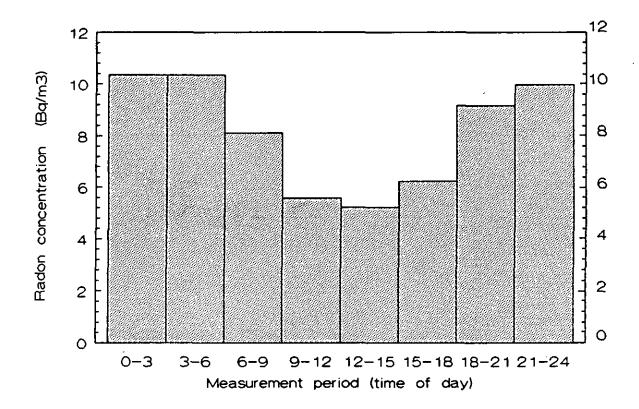


Figure 3,2. Diurnal variation of outdoor ²²²Rn concentration at Chester (New Jersey, USA), showing the averages of three-hour data from 1977-1982 (UNSCEAR, 1988).

air current in the early morning which carries radon along (Wilkening, 1990). Consequently, the difference between the highest and lowest concentrations is smaller on cloudy days.

The behaviour of the ²²²Rn daughters in the atmosphere does not differ essentially from that of their parent, apart from the influence of wet and dry deposition (see 3.1.2) (Dueñas et al., 1990).

Since ²²²Rn exhalation from sea water is about two orders of magnitude less than from soil (see 2.2), there is a clear difference between the concentration in continental air and marine air. Concentrations below 1 mBq m⁻³ have been measured in air over the open oceans. The normal range in marine air is about 1-100 mBq m⁻³ (UNSCEAR, 1982). This means that wide variations can occur in the ²²²Rn concentration at the transition from sea to land. It is obvious that the ²²²Rn concentration in air near the coast depends on the direction of the wind. The concentrations are low when the wind blows from the sea and relatively high when it blows from the land. The influence of continental air on the ²²²Rn levels in air over the sea is still noticeable at distances of up to several thousands of kilometres from the coast, as has been observed in ²²²Rn measurements on Hawaii (UNSCEAR, 1982).

The average ²²²Rn concentration in air above continental areas is about 9 Bq m⁻³. Since coastal regions are more densely populated and have lower ²²²Rn concentrations, the population-weighted world average of the ²²²Rn concentration- is about 5 Bq m⁻³ (UNSCEAR, 1988). Combining this value with an estimated average equilibrium factor F (see 1.2.2) of 0.8 at 1 m above ground (UNSCEAR, 1988) yields an average equilibrium equivalent ²²²Rn concentration of about 4 Bq m⁻³. The normal range, excluding extreme values, is from 1 to 10 Bq m⁻³.

Radon-222 is used as a natural radiotracer in studies of continental and local air dispersion, of local air pollution such as, for example, in urban areas (UNSCEAR, 1982; Cothern and Smith, 1987), and of vertical diffusion in the atmosphere, where ²²²Rn is the most widely used natural tracer (Dueñas et al., 1990).

3.1.2 <u>Deposition of radon daughters</u>

As has already been reported in 1.2.2, radon is generally not in radioactive equilibrium with its daughters in the atmosphere. The degree of radioactive equilibrium depends to a large extent on meteorological conditions (UNSCEAR, 1988) because, unlike the noble gases ²²²Rn and ²²⁰Rn, their daughters readily attach to aerosol particles owing to their chemical reactivity and charge. Prior to attachment, clustering of free daughter products occurs. The behaviour of attached daughters is determined chiefly by the physical properties of the carrier aerosol, such as the diameter. These aerosols are removed from the atmosphere by dry and wet deposition. The occurrence and behaviour of the relatively short-lived daughters of ²²²Rn (²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi and ²¹⁴Po) is highly dependent on the buildup from ²²²Rn and radioactive decay. The physical half-lives are short compared to the time scales of the atmospheric removal mechanisms, with the exception of washout (UNSCEAR, 1982).

Particularly during longer periods of dry stable weather with a light offshore breeze, there is much buildup of 222 Rn activity in the atmosphere and sufficient time for daughter products to attach to aerosols. When this is followed by heavy rain or snowfall, there is a greatly increased deposition of daughters above land masses due to washout (Smetsers, 1990). As a consequence, the dose equivalent rate in air from gamma radiation at ground level increases greatly. This is illustrated in Figure 3.3 for the Kollumerwaard location. Since particularly both 214 Pb ($t_{1/2} = 27$ min) and 214 Bi (20 min) are responsible for this gamma radiation, and the parent nuclide 218 Pb has a much shorter half-life (3.1 min), the dose equivalent rate in air from gamma radiation decreases again rapidly after rinsing the atmosphere (apparent half-life about 0.5 hour).

The world-averaged concentration of the long-lived 222 Rn daughter 210 Pb ($t_{1/2}=21$ years) in rainwater is about 0.07 Bq 11 , which, assuming an annual precipitation of 1000 l 10 m⁻², corresponds to an annual average wet deposition of 70 Bq 11 m⁻² (Eisenbud, 1987). The average deposition rate above land is greater than above the sea. In the Netherlands, the measured annual wet and dry deposition of 210 Pb is about 100 Bq 11 m⁻² (van Sonderen et al., 1990). Because of the relatively long half-life of 210 Pb, accumulation of deposited 210 Pb occurs. The equilibrium value for the accumulated 210 Pb deposit is about 3000 Bq 11 m⁻². This value also applies to the daughter product 210 Po (11 2 = 138 days) formed from this 210 Pb.

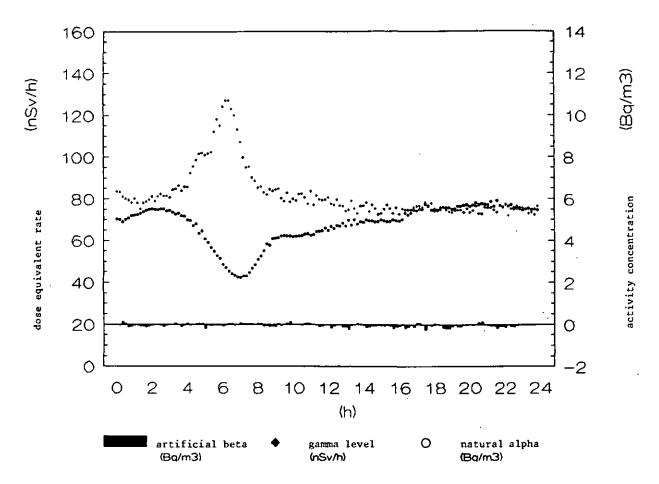


Figure 3.3. Increase in the dose equivalent rate in air from gamma radiation at ground level during a period of snowfall. Results for the Kollumerwaard location from the National Radioactivity Monitoring Network in the Netherlands.

3.2 BEHAVIOUR IN SOIL

3.2.1 Movement in soil

Movement of radon in soil is caused by diffusion and convection, which are both influenced by physical characteristics of the soil, such as permeability and porosity (see 1.1.2). The type of soil and the moisture content are therefore important factors for radon exhalation. Radon exhalation from soils with very high or very low moisture content is relatively low, but reaches a peak at an intermediate water content of about 25% (Stranden et al., 1984). Consequently, the water table is one of the factors determining the exhalation rate. In addition, the presence of cracks and breaches in the earth affects the supply of radon from deeper layers of the earth. Along geological fault lines, radon can be transported via gas or water movements from rocks deep inside the earth, resulting in much larger rock masses acting as a source and local "222Rn sources" can arise. A well-known example in the USA is the Reading Prong formation in Pennsylvania. Radon-222 concentrations of up to 100,000 Bq m⁻³ have been measured in homes in this area (Nero et al., 1990).

The convective flow from the soil is determined primarily by the pressure difference between surface air and soil gas. When the atmospheric pressure increases, the exhalation of radon is slowed. Conversely, when the air pressure decreases, soil gas is sucked from the soil, the result being that the exhalation rate can increase considerably (Miles and Algar, 1988). Rocks deep inside the earth act as a ²²²Rn source during the extraction of groundwater and minerals such as natural gas (see 2.3.3) and petroleum. The same is true for the extraction of steam from high-temperature rocks deep inside the earth for the production of geothermal energy (UNSCEAR, 1988). A schematic representation of the principal processes and parameters in ²²²Rn production and migration in soil is presented in Figure 3.4.

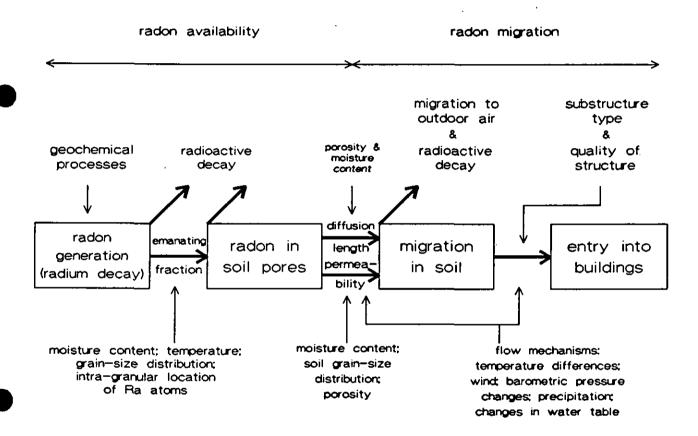


Figure 3.4. Schematic representation of ²²²Rn production and migration in soil and its entry into buildings (From Nazaroff and Nero, 1988).

3.2.2 Entry into dwellings

Diffusion and convection are also the two principal mechanisms by which radon in soil gas is transported into houses and other buildings. The rate of diffusion is determined by the properties of the building materials used, such as porosity and moisture content, and the thickness of the barriers between the ground and the living space of the building. The convective flow from the soil into the house is caused by pressure differences and is therefore also determined by "leaks" in the building structure, such as holes, gaps, and cracks caused by shrinkage resulting from ageing. In addition, building construction practices, wind direction, temperature and the habits of the occupants (e.g. with regard to ventilation and heating) play a role. Convective flow from soil is probably responsible for

the relatively high ²²²Rn levels in some homes (Eisenbud, 1987). Because of the high concentrations in soil gas (20,000 Bq m⁻³ being a worldwide typical value) (Nazaroff and Nero, 1988), a small-supply of soil air is already sufficient for the soil to have a dominant influence.

The primary route for radon entry by diffusion is through the entire floor slab. In addition, there may be a route via cavity walls and around service pipes, so that radon is also directly transported from the soil to the upper floors by diffusion. Although the diffusion rate can vary owing to the influence of the moisture content this process nevertheless has more the character of a "static" radon source (de Meijer, 1991).

Convective transport of radon from soil into a house is caused by pressure differences between indoor air and soil gas. As in the case of exhalation from soil, the principal factor here forms the fluctuations in air pressure (see 1.1.3). A drop in pressure sucks radon from the soil into the house. Pressure differences are also induced by winds and temperature differentials. If there is wind, the pressure on the windward side of the house is greater and that on the leeward side less than the free-stream pressure in the open air. In principle, this effect also occurs with respect to soil gas, because, depending on the permeability of the soil, the pressure at the surface of the soil is rapidly (sandy soils) or slowly (clayey soils) propelled through the soil gas (see Table 1.2). A pressure differential also exists across a vertical wall separating air masses of different temperature. It varies with height and is approximately proportional to the temperature difference, giving rise to a convective flow from the soil into the house. When it is warmer indoors than outdoors, the air pressure indoors falls in comparison with the outdoor air pressure, which sucks radon-bearing soil gas into the house. This is known as a "stack effect" (Nazaroff and Nero, 1988; Loos, 1989). Clearly, this effect depends on the heating practices of the occupants which is, of course, affected by the season. Because of the occurrence of rapid variations in convective flows, particularly with a sudden change of air pressure, convection has been described as a "dynamic" radon source (de Meijer, 1991).

A number of compartments and partitions can be involved in radon transport from soil into a house by diffusive as well as by convective flows. The free spaces between the floor slab and the soil, and/or the crawl spaces and/or the cellar are often important "intermediaries" between the soil and the house. The intermediary role of the crawl space is further enhanced by the fact that the pressure in the living room is usually lower than in the crawl space (Put and de Meijer, 1989). The possibly positive influence of the crawl space deserves mention here because this understructure type may lead to lower exposures than do other types.

Transport from soil into a house via the cavity wall can also be important. The principal entry pathway in houses with normal ²²²Rn concentrations is usually not known. Investigations in test houses have not yet given a decisive answer about the relative importance of the cavity wall as an entry pathway.

There are also a number of indirect routes for ²²²Rn transport from the soil into the house. Well-known examples are tap water and natural gas (see 2.3 and 2.6). In addition, transport via sewer pipes is possible. It is true that the water seals in the pipes prevent ²²²Rn supply by convective flows but diffusion can be important. Moreover, a much larger part of the soil can act as an ²²²Rn source via the sewerage system. Relevant information

is not available. Figure 3.5 presents an overall picture of the sources and entry routes of ²²²Rn in the indoor environment.

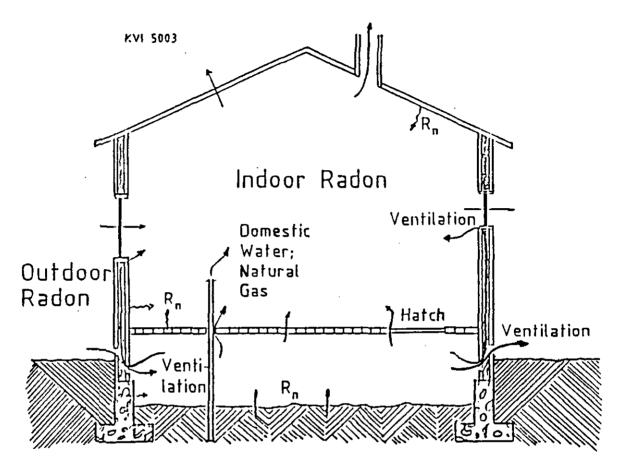


Figure 3.5. Summary of the sources and entry routes of ²²²Rn in the indoor environment (Loos, 1989).

3.3 DISTRIBUTION OF RADON DAUGHTERS IN ORGANISMS

There is a limited number of examples of "bioaccumulation" of ²¹⁰Pb and/or ²¹⁰Po derived from ²²²Rn in the atmosphere. A well-documented case is that resulting from the deposition of airborne ²¹⁰Pb and ²¹⁰Po on reindeer moss. Reindeer and caribou graze on this lichen in the winter (UNSCEAR, 1988). Because these animals eat large amounts of this herbage, their meat, which is the main source of food of tens of thousands of people, contains unusually high concentrations of ²¹⁰Pb. This meat is rarely eaten in the Netherlands. Lead is a bone seeker and, consequently, the ²¹⁰Pb consumed by these animals accumulates in their bones.

The intake of ²¹⁰Pb and ²¹⁰Po by grazing cattle does not lead to high concentrations in meat and milk relative to other foodstuffs (Smith-Briggs and Bradley, 1984). In view of the high consumption of milk (products), however, milk, in addition to other beverages, bread and cereal products, belongs to the foodstuffs which account for about 60% of the daily intake of these radionuclides by humans (Smith-Briggs and Bradley, 1984).

A second example of an organism in which ²¹⁰Po accumulation occurs is the marine mussel. Mussels are very efficient accumulators of trace elements, including polonium,

from seawater (Köster et al., 1985). High concentrations are observed in mussels, especially in the vicinity of non-nuclear industries which discharge ²¹⁰Pb and ²¹⁰Po to air and water. The issue of technologically enhanced concentrations of radionuclides is beyond the scope of this integrated criteria document, because these nuclides are not derived from ²²²Rn in the atmosphere but from radium-bearing raw materials used in the processing industry.

Tobacco leaves contaminated with ²¹⁰Pb due to atmospheric deposition is a significant source of exposure for smokers. The ²¹⁰Po formed from this ²¹⁰Pb and the deposited ²¹⁰Po are directly taken up by tobacco leaves. This route of contamination is probably more important to the tobacco plant than direct uptake from soil (Eisenbud, 1987; Cothern and Smith, 1987). Although there is no consensus on the magnitude of the resultant additional doses to smokers, doses clearly are relatively large (Eisenbud, 1987; NCRP, 1987). The NCRP in particular has advocated more research into the lung doses from the "consumer product" tobacco (NCRP, 1987).

3.4 SUMMARY AND CONCLUSIONS

Radon exhalation from the ground is the main source of radon in the atmosphere. Its dispersion in the atmosphere depends upon metereological factors, such as vertical temperature gradient, wind direction, wind speed, and vertical turbulent mixing. Radon-222 rises much higher into the atmosphere than ²²⁰Rn. There are diurnal and seasonal patterns of variation in the ²²²Rn concentrations at ground level. In addition to radioactive decay, wet and dry deposition are important mechanisms for removing ²²²Rn decay products from the atmosphere. There are large differences between the concentration of ²²²Rn in continental and marine air. The population-weighted world average of the ²²²Rn concentration in outdoor air is 5 Bq m⁻³. The average value and typical range for the equilibrium equivalent ²²²Rn concentration are about 4 Bq m⁻³ and 1-10 Bq m⁻³.

Movement of radon in soil is caused by diffusion and convection. Key parameters in determining the radon exhalation rate from soil are soil type, moisture content and the water table. Radon entry from soil gas into houses also occurs through diffusion and convection. Convective flow is strongly influenced by pressure differences. Consequently, atmospheric pressure changes, wind strength and temperature differences are important parameters for convective transport from soil. The crawl space is important in the transport of radon from soil into a building. The cavity wall is possibly also an entry pathway. Mention should be made of the possibly favourable role played by the crawl space. Radon movement from the soil into the house may be less in houses with a crawl space than in houses without a crawl space.

Mussels are an example of organisms which accumulate ²¹⁰Po present in the marine environment. Smokers receive a relatively high dose from inhalation of the ²¹⁰Po contained in tobacco leaves, which is presumably derived from ²²²Rn in the atmosphere.

4 MEASUREMENT TECHNIQUES

4.1 AIR

Various instruments and techniques have been established for measuring the concentration of ²²²Rn in air. The National Council on Radiation Protection and Measurements (NCRP, 1988) and Nazaroff (in Nazaroff and Nero, 1988) have surveyed the literature in this field. The techniques may be divided into three categories: instantaneous, continuous and integrating techniques. The first two categories are used almost exclusively in research projects, i.e. as a method for making single concentration measurements and for determining the variation of the concentration with time, respectively. Integrating techniques are used for the determination of indoor ²²²Rn concentrations. The methodologies for measuring ²²⁰Rn are treated in 4.1.4.

4.1.1 <u>Instantaneous measurements</u>

Measurement of the instantaneous concentration of ²²²Rn in air is most commonly made with a system consisting of a scintillation cell where the interior surface is coated with a scintillator, usually zinc sulphide, and a photomultiplier tube which detects the light pulses produced. The detection limit of this technique is about 10 Bq m⁻³ for a 100 cm³ cell and a 60-minute counting period (Lucas, 1957). Since the first description by Lucas, other cell designs have been proposed in the literature, aimed to increase the counting efficiency and the sensitivity (NCRP, 1988).

Alternatives to the "Lucas" cells are used on a limited scale. One example is the ionization chamber, with which the existence of ²²²Rn was first demonstrated. Other, less costly instruments have virtually replaced ionization chambers, with the exception of their use for accurate laboratory measurements.

Another technique for the instantaneous measurement of ²²²Rn in air is known as the "two-filter" method. It uses a large chamber with a filter at each end. Sample air is pumped through the chamber. The inlet filter removes all radon daughters. Radon gas passes through, and inside the chamber a small fraction decays. The decay products are deposited on the outlet filter, which after quantification, is used as a measure of the radon concentration. This technique has been employed, for example, within the SAWORA research programme (Groen et al., 1986). Because of the large volume of the chamber (120 l in the study mentioned), this technique is only suitable for laboratory use.

4.1.2 (Semi-)continuous mesurements

It is known that the indoor radon concentrations are subject to diurnal and seasonal variations (see 5.4.1). Consequently, single instantaneous measurements are of only limited value for estimating the radiation risk from ²²²Rn. A follow-up period, of the order of several months, is necessary if a statement with a certain degree of confidence on annual mean concentrations in buildings is to be made. Two types of measurements can be used for estimating the annual average concentration: (semi-)continuous measurements of ²²²Rn in time and an integrated measurement. The latter will be discussed in 4.1.3. In the semi-continuous measurements, air samples are collected batchwise (for example, every half-hour), and the time interval is used to fill the measurement chamber and to analyze the sample. For both the semi-continuous and continuous measurements, active sampling

in which the air is introduced into the measurement chamber by means of a pump is generally employed. This sampling method has several drawbacks (expensive, not maintenance-free and too noisy), so that its application has been limited to research projects.

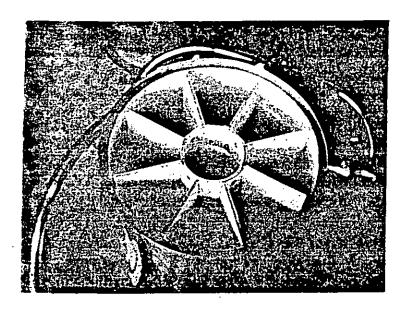


Figure 4.1. A flow-through scintillation cell for measuring the radon concentration in air (KVI).

A common method, with which already much experience has been gained, is the flow-through Lucas flask technique (see 4.1.1). The detection limit reported for a 3 1 scintillation flask (see Figure 4.1) was 10 Bq m⁻³ after a counting time of 30 minutes (Stoop et al., 1991). In addition to the Lucas cell, portable ionization chambers with specially designed electrodes are used. Baltzer et al. (1989) reported that these devices are capable of measuring a concentration of 150 Bq m⁻³ within one hour with a precision of 10%. Under the same conditions, Katase et al. (1986) claim a detection limit of about 20 Bq m⁻³. Furthermore, these ionization chambers distinguish between different alpha energies, so that in addition to ²²²Rn, ²²⁰Rn and several daughters can also be quantified. Another principle is based on the electrostatic collection of ²¹⁸Po (and ²¹⁴Po) on a film of Mylar. The voltages applied vary widely. The flow-through instruments generally use a surface barrier detector for alpha detection (Watnick et al., 1986).

One drawback of all flow-through instruments described here is that radon daughter products deposit on the cell walls, resulting in increased background count rates. Various methods have been described for the corrections required in the counting results for the contribution of the deposited daughters, (Busigin et al., 1979; Thomas and Countess, 1979). It will be clear, however, that the corrections reduce the precision of the analyses.

4.1.3 <u>Integrating techniques</u>

Integrating techniques have been (and are) widely used in a variety of monitoring programmes. The etched-track detector in a cup developed in Karlsruhe (Urban and Piesch, 1981) has been used for the determination of the annual average ²²²Rn concentration in the Netherlands. It is an example of passive sampling, where the ²²²Rn atoms enter the sensitive volume by diffusion. The emitted alpha particles leave damage tracks on the plastic detector taped to the bottom of the cup. The tracks become visible upon etching, and are then counted under a microscope. The number of tracks produced over the exposure period is a measure of the quantity of ²²²Rn that has diffused into the cup. The integrating time is ordinarily 3-12 months. The detection limit is of the order of 5-10 Bq m⁻³ for a four-month exposure period.

Another technique, which is popular, especially in the U.S.A., is based on the ability of activated carbon to adsorb ²²²Rn. Following exposure, the quantity of daughters formed from the decay of adsorbed ²²²Rn is measured by gamma spectrometry (see 4.2.1) or by liquid scintillation counting (Prichard and Mariën, 1983). The charcoal adsorption procedure is simple but not very accurate. The disadvantages include:

- The degree of radon adsorption is greatly influenced by humidity changes;
- Because of the short half-life of ²²²Rn and its daughters, the exposure period cannot usefully be longer than 2-7 days. This integration period is too short for drawing firm conclusions about the annual average concentration;
- Calculation of the true average over the sampling period is difficult because, given the changing ²²²Rn concentrations (see 5.4.1), the adsorption will not be linear with time.

Another relatively widely used technique is based on the use of a thermoluminescent dosimeter (TLD). In this method, as described by George and Breslin (1977), the charged ²¹⁸Po and ²¹⁴Po ions formed from the decay of ²²²Rn are collected on a Mylar film maintained at -900 V potential. A TLD is positioned behind the Mylar film. The lower limit of detection is about 1 Bq m⁻³ of ²²²Rn for a one-week sampling period. Cowper and Davenport (1978) give a similar detection limit. Mariello and Harley (1987) have replaced the batteries for applying the collecting potential with an electret. This is a sheet of Teflon foil which retains its charge for a long period. The electret was charged to -1200 V. The result is a considerably smaller and less expensive monitor. The latest developments in this field point to the use of an electret without a TLD (Kotrappa et al., 1990; Summers et al., 1990). The drop in surface voltage of the electret over a known period is a measure of the number of ions formed within the sensitive volume during that time interval which can be converted into ²²²Rn concentrations. The lower limit of detection was about 10 Bq m⁻³ for a three-month exposure period (Kotrappa et al., 1990).

In addition to the desired detection limit, the cost aspect also plays a role in deciding which system to choose. For the etched-track detector in a cup used by the KVI, the cost is Dfl. 120.- per analysis (four-month exposure period). The activated carbon technique costs Dfl. 50.- to Dfl. 100.- per measurement. No experience has been gained in the Netherlands with the other methods. However, the costs of analysis are probably considerably lower. A summary of the techniques for measuring radon in air is given in Table 4.1.

Table 4.1. Summary of methods for measuring 222Rn in air

Method (sampling period)	System	Detection limit* Bq m ⁻³	Reference
Instantaneous measurement (1-60 min)	Lucas cell Two-filter method	10 0.6	Lucas (1957) Groen et al. (1986)
(Semi-)continuous measurement	Flow-through Lucas cell	10	Stoop et al. (1991)
(10-120 min)	Ionization chamber	<20	Katase et al. (1986)
Integrated	Etched-track detector	5-10	Put (pers.comm.)
<pre>measurement (1-12 months)**</pre>	Activated carbon canister	10	George (1984)
•	TLD	1	George & Breslin(1977)
	Electret	10	Kotrappa et al. (1990)

^{*} For particulars such as sensitive volume and counting time, the reader is referred to the text.

4.1.4 Techniques for the measurement of ²²⁰Rn

The techniques for measuring ²²⁰Rn in air may also be divided into the three aforementioned categories: instantaneous, continuous, and integrating techniques. An example of each category is given below.

The first system for measuring ²²⁰Rn was described by Israel and Israel in 1965 (NCRP, 1988). It used two large ionization chambers and a decay chamber positioned in between. Air was drawn into the first ionization chamber where the combined ²²²Rn and ²²⁰Rn activities were recorded. The air then passed through the decay chamber with a sufficiently long transit time for most of the ²²⁰Rn to decay leaving only ²²²Rn to be measured in the second ionization chamber. The ²²⁰Rn concentration was obtained from the difference between the two readings.

In addition to ²¹⁸Po and ²¹⁴Po, an electrostatic field also collects ²¹⁶Po, a decay product of ²²⁰Rn. If a suitable detector is used (for example, a surface barrier detector), the concentration of ²¹⁶Po can be determined separately (Folkerts et al., 1984).

A passive etched-track technique for ²²⁰Rn has been described by Somogyi et al. (1984). Each cup has two plastic detectors. The first measures both ²²²Rn and ²²⁰Rn and the second, positioned further away from the inlet, ²²²Rn only.

4.2 BUILDING MATERIALS

4.2.1 Gamma spectrometry

Gamma spectrometric analysis of building materials is generally performed on the crushed material (≤ 1 cm diameter) using a Ge(Li) or a Ge detector. On the basis of the measured concentrations of ²¹⁴Pb and ²¹⁴Bi on the one hand, and ²¹²Pb and ²¹²Bi on the other, the concentration of ²²⁶Ra (²²²Rn) and ²²⁴Ra (²²⁰Rn), respectively, can be determined, assuming radioactive equilibrium exists (Ackers, 1985).

^{**} The exposure period for the activated carbon technique is 2-7 days.

4.2.2 Exhalation rate

A large number of methods for measuring the exhalation rate from building materials has been described in the literature. Broadly, they can be divided into two types. The first method consists of placing a test wall of the building material to be examined in a closed chamber and measuring the ²²²Rn concentration in the free volume of the chamber after a pre-selected time (Mustonen, 1984; Poffijn et al., 1984; Stranden et al., 1984). This method has the disadvantage that the exhalation rate decreases with increasing ²²²Rn concentration in the free volume of the chamber, due to back diffusion. In practice, radon is exhaled into a virtually radon-free atmosphere, resulting in the so-called free exhalation rate. This method can be improved by following the buildup of radon activity in the chamber as a function of time. The free exhalation rate is then determined from the initial growth rate (Jonassen, 1983; Folkerts et al., 1984; Ulbak et al., 1984). In addition, the effect of back diffusion can be reduced by making the ratio of chamber volume to sample volume greater than 10, as suggested by Poffijn et al. (1984). The difference between the observed exhalation rate and the free exhalation rate is then less than 5% (Samuelsson, 1987).

The effect of back diffusion can also be virtually eliminated by continuously ventilating the above-mentioned chamber. This so-called purge-and-trap method has been described by van Dijk and de Jong (1989) and leads to a deviation from the free exhalation rate of a maximum of 0.5% (222Rn). This is a convenient method for investigating the effect of external parameters such as air-exchange rate and humidity on the exhalation rate.

In the second method, a hemispherical chamber is sealed to the surface of the material to be examined (Folkerts et al., 1984; Ackers et al., 1985; Aldenkamp et al., 1991), and the growth of the radon concentration inside the chamber is then continuously measured. Therefore, this method is essentially the same as the method described above, but differs in that it may be used on an existing wall. However, a major disadvantage of this method is that it is not always so easy to seal the hemispherical chamber hermetically to the building material when this has a rough surface. Furthermore, a proportion of the accumulated radon may leak to the outside air via the pores in the material, especially when this has a high porosity. A typical exhalation meter is shown in Figure 4.2.

In addition to a division by sampling method, the methodologies described in the literature may be further subdivided according to the detection technique employed. Since these techniques do not differ from those described in Section 4.1, the reader is referred to this section.

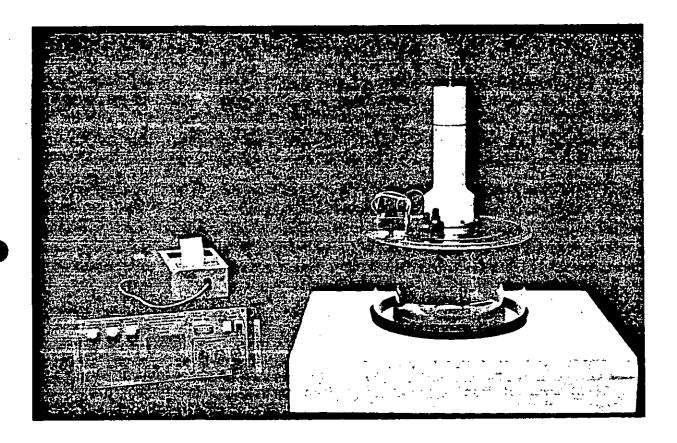


Figure 4.2. A radon exhalation meter for building materials (TNO).

4.2.3 Emanation coefficient

Only a fraction of the ²²²Rn and ²²⁰Rn atoms produced by decay in a particle of the building material is released into the pore volume of the material. This fraction is known as the emanation coefficient, and has been defined in 1.2.3. If it is assumed that all pores of the material are in contact with the open air, the emanation coefficient for ²²²Rn can be calculated from the measured exhalation rate from the building material under consideration (van Dijk and de Jong, 1989):

$$\eta = E / (0.5 d \rho \lambda A) \tag{4.1}$$

where

 η = the emanation coefficient

E = the exhalation rate of 222 Rn (Bq m⁻² s⁻¹)

0.5 d =the half-thickness of the wall (m)

 ρ = the density of the building material (kg m⁻³)

 λ = the decay constant of ²²²Rn (2.1x10⁻⁶ s⁻¹)

A = the 226 Ra concentration (Bq kg $^{-1}$)

To determine the emanation coefficient, Ackers (1989) used crushed building material and measured the ²²²Rn released from it with a zinc sulphide detector. These two methods

yielded similar emanation coefficients. For example, values of 0.22 and 0.20, respectively, were found for nitrogypsum (de Jong and Ackers, 1990).

4.3 SOIL

4.3.1 Gamma spectrometry

Gamma spectrometric analysis of soils is performed in the same way as described in 4.2 for building materials. The results are usually expressed on a dry weight basis, so that the dry weight of the soil must also be determined.

4.3.2 Exhalation rate

Unlike the exhalation rate from building materials, the radon flux from soil must always be measured "in the field". Methods used in measuring radon flux are shown schematically in Figure 4.3 (Wilkening, 1990).

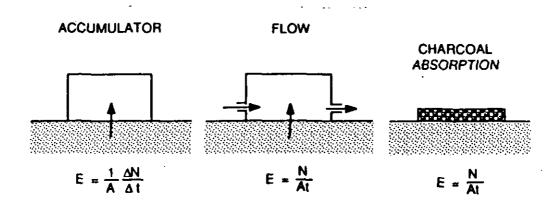


Figure 4.3. Methods used in measuring radon flux, E. A is the area being measured, t is the accumulation time and N is the quantity of radon.

The simplest accumulation method consists of sealing the open end of a vessel to the surface being measured. The exhalation rate can then be determined from the increase in the radon concentration measured over a given time period. This technique has been discribed by such authors as Ackers (1985a). The radon concentration in the vessel increases linearly with time only when certain requirements are met (Wilkening, 1990). For example, the concentration in the vessel must remain sufficiently low to prevent back diffusion (see also 4.2.2).

The flow method has the advantage that it is capable of simulating a natural situation. Several variations of the flow method, differing mainly in the detectors employed, have been developed. In the original flow method, the air in the accumulation vessel was continuously circulated through a charcoal trap which collected the radon. The radon was then measured by gamma spectrometry. The current flow-through accumulators employ continuous monitoring of radon in the outlet airstream. The measuring instruments used have already been discussed in 4.1.2.

Countess (1976) used a charcoal gas-mask canister as the accumulator. After a given exposure period, the entire canister was measured in a gamma spectrometer. Countess used an NaI scintillation detector to measure gamma radiation from both the ²¹⁴Pb and ²¹⁴Bi radon daughters. The canister procedure is simple, but not very accurate (see also 4.1.3). However, it is still the chosen method for performing large numbers of measurements.

4.3.3 Soil gas

The same methods are used for the determination of the radon concentration in soil gas as previously decribed for air (4.1). For example, Åkerblom et al. (1984) used both activated charcoal and etched-track detectors in their study. The detectors were placed 1 m below the soil surface, and exposed for 4-8 days and 3-6 weeks, respectively. Because of the high concentrations of radon in soil gas, a few tens of kBq m³ (see 5.1), detection problems do not arise.

4.4 WATER

Techniques for measuring ²²²Rn in water exploit the fact that radon is a gas. The radon can be released from the sample to be analyzed by passing a carrier gas, for example nitrogen, through the sample. When activated carbon is used to collect the ²²²Rn, the ²²²Rn is removed from the carbon after the collection period by heating and transferred to a Lucas cell for counting (Prichard et al., 1980; Poffijn et al., 1984). This measurement technique has already been described in 4.1.1. The detection limit is about 1 mBq of ²²²Rn. A rapid method, in which the ²²²Rn is collected on silica gel, has been described by Darrel (1973). It employs a liquid scintillation counter. The detection limit is about 10 mBq of ²²²Rn (van Dijk and de Jong, 1989).

A survey technique for ²²²Rn in water has been described by Prichard et al. (1980). The sample to be measured is analyzed directly for ²²²Rn content by liquid scintillation counting.

4.5 STANDARDIZATION OF MEASUREMENT TECHNIQUES

As part of the "Normstar" project, a large number of pre-standard have been issued in the field of radioactivity and radiation measurements (van den Ende and van Lith, 1988). Since the project's emphasis was on the standardization of measurement techniques which are used after a nuclear accident, only a small number have a bearing on the methods described in this chapter. One example is the pre-standard NVN 5623 (Radioactivity measurements. Determination of the activity of gamma radiation-emitting nuclides in a sample by solid-state gamma spectrometry).

The Netherlands Standards Institute (NNI) has recently organized a workshop on the content of a possible follow-up project "Normstar II". Various parties have called for inclusion of measurement techniques for radon in the programme (NNI, 1991).

4.6 SUMMARY AND CONCLUSIONS

The literature on the measurement of radon is very extensive. The major techniques have been briefly discussed in this chapter. The techniques for measuring ²²²Rn in air have been divided into three categories: instantaneous, (semi-)continuous and integrating techniques. The last category is particularly suitable for application to dwellings and other buildings. The etched-track detector in a cup for ²²²Rn used in the Netherlands is satisfactory and has a sufficiently low detection limit after four months of exposure. The technique is less suitable for shorter exposure periods and moreover too expensive for screening purposes. Less costly alternatives, based on thermoluminescent dosimeters (TLD) and electret exist, but they have not been tested under Dutch conditions (low concentrations).

For the other matrices (building materials, soil, water) reliable methods for ²²²Rn are also available in the Netherlands. In contrast, the number of methods for ²²⁰Rn is limited. To date, the measurement techniques have not been standardized.

5 <u>CONCENTRATIONS AND EXPOSURE LEVELS</u>

5.1 CONCENTRATIONS IN SOIL

The average worldwide activity concentration of ²²⁶Ra and ²³²Th in soil is taken to be 25 Bq kg⁻¹, with a typical range of 10-50 Bq kg⁻¹ (UNSCEAR, 1988). Investigations of the activity concentrations of these nuclides in Dutch soils yielded average values of 25 and 35 Bq kg⁻¹, respectively. Table 5.1 presents a compilation of the Dutch data.

Table 5.1. Activity concentrations (0-20 cm) of a few Dutch soil types (Ackers, 1985; Bannink et al., 1986)

Soil description	²²⁶ Ra	²³² Th	
	(Bq kg ⁻¹)	(Bq kg ⁻¹)	
River clay	20 - 50	20 - 80	
Marine clay	25 - 40	30 - 50	
Dune sand	5	10	
Windborne sand deposit	10 - 15	10 - 30	
Loess	15 - 40	30 - 60	
Peat	5 - 30	10 - 60	
Average value (rounded off)	25	35	

The emanation coefficient (see 1.2.3) is highly dependent on the type of soil. In his review Wilkening (1990) gives a range of 0.2-0.7, with an average of 0.4. UNSCEAR (1988) has adopted a range of 0.01-0.8, with 0.2 as a typical value. This parameter has not been measured in the Netherlands. The moisture content of the soil plays an important role in determining the ²²²Rn concentration in soil gas. The explanation for this lies in the large difference between the diffusion coefficient of ²²²Rn in water and in air (see 1.1). A high water table will therefore tend to reduce the concentrations in soil gas and in the air above the soil. This effect has been studied in more detail by Loos et al., (1991). Their data indicate that a correlation exists between the average ²²²Rn concentration and the average groundwater level in the preceding week. Generally, for the measurement location under consideration, the ²²²Rn concentration in soil gas increased by 1000 Bq m⁻³ for every 10-cm fall in the water table.

Near the test house in Roden, ²²²Rn concentrations of up to 8000 Bq m⁻³ were observed in soil gas at a depth of 40 cm (Loos et al., 1991). It will be clear that the concentration in soil gas is highly dependent on the radium content of the soil and the soil porosity. The ²²⁶Ra concentration in the soil studied is relatively low, about 10 Bq kg⁻¹. The ²²²Rn concentration in soil gas, a few metres below ground level is on average 50,000 Bq m⁻³ (UNSCEAR, 1988). Under influence of the difference between the ²²²Rn concentration in soil gas and outdoor air or a pressure gradient an upward flow is created. Table 5.2 summarizes the exhalation rates from the soil surface.

Table 5.2. Exhalation rates of 222Rn from the soil surface

Reference	Range (mBq m ⁻² s ⁻¹)	Average (mBq m ⁻² s ⁻¹)	
Ackers (1985a)	0.3 - 30	nr	
Ackers (1988)	0.2 - 11	4	
NCRP (1988)	nr	17	
UNSCEÀR (1988)	0.2 - 70	16	
Wilkening (1990)	5 - 30	19	

nr = not reported

The Dutch measurements (Ackers, 1985; 1988) were carried out at ten locations. Clayey soils generally show the highest exhalation rates (average 7 mBq m⁻² s⁻¹). Those for sandy and peat soils are, on average, about 3 mBq m⁻² s⁻¹ (Ackers, 1988). Table 5.2 shows that the average exhalation rate in the Netherlands is considerably lower than in other countries. Wilkening (1990) reported that the exhalation rate of ²²⁰Rn (1.5 Bq m⁻² s⁻¹) is a factor of 100 higher than that of ²²²Rn.

5.2 CONCENTRATIONS IN BUILDING MATERIALS

By comparison with other countries, much research has been carried out in the Netherlands into the contribution of building materials to the total radiation dose resulting from "Building construction and occupancy". It concerns studies on the common, conventional building materials, as well as on (future) building materials in which industrial by-products have been incorporated as substitutes for natural products. Table 5.3 gives a few data on the principal building materials in use in the Netherlands.

Table 5.3. A few parameters of Dutch building materials (Ackers, 1985; 1985a; 1989; Van Dijk and De Jong, 1989)

Building material	Activity con ²²⁶ Ra (Bq kg ⁻¹)	centration ²³² Th (Bg kg ⁻¹)	Emanation coefficient	Exhalation rate of ²²² Rn (mBq m ⁻² s ⁻¹)
Concrete	10 - 20	10 - 25	0.10 - 0.25	0.5 - 1.0
Aerated concrete	15 - 25	5 - 10	0.15 - 0.25	0.2 - 0.3
Brick Sand-lime brick	30 - 50 5 - 10	30 - 50 5 - 10	0.005 - 0.03 0.16	0.05 - 0.2
Natural gypsum Tiles/paving tiles	1 - 10 40 - 90	1 - 5 50 - 80	0.10 - 0.20 0.001 - 0.02	<0.01 - 0.15

Table 5.3 shows that there is not much difference between the concentrations of ²²⁶Ra and ²³²Th in these building materials, with the exception of brick. Folkerts et al. (1984) determined the exhalation rate of ²²⁰Rn as well as of ²²²Rn, and found it to be approximately a factor of 100 higher. However, applying a finishing coat to the building material under consideration will greatly affect the magnitude of precisely this parameter (van Dijk and de Jong, 1989). Industrial by-product materials generally have higher ²²⁶Ra and ²³²Th concentrations than natural materials. A number of these by-products are listed in Table 5.4.

Table 5.4 Activity concentrations of two radionuclides in industrial by-product materials (Ackers, 1985; 1989; van Dijk and de Jong, 1989; de Jong and Ackers, 1990)

By-product	Activity co	Emanation coefficient	
	(Bq kg ^{.1})	(Bq kg ⁻¹)	
Fly ash	100 - 300	80 - 300	0.01 - 0.04
Blast-furnace \$lag	15 - 120	20 - 250	0.01 - 0.04
Phosphogypsum	500 - 800	2 - 10	
Nitrogypsum	30	20	0.08 - 0.2
Flue gas-desulphurization gypsum	3	<4	

Their incorporation into building materials makes a significant contribution to solving the waste problem, and also saves primary raw materials. Besides, policy is aimed at ensuring that the radiation dose received by the occupants of dwellings due to building materials will not increase (standstill principle). As part of the RENA research programme, an exploratory study has been made on the usability of various by-products, including phosphogypsum, nitrogypsum, blast-furnace slag and fly ash (Winder et al., 1990). It concluded that the use of phosphogypsum does not fulfill the aforementioned standstill principle (see 1.3.3). However, its use as gypsum board does not lead in practice to a detectable increase in the indoor airborne radon concentration (Veermans et al., 1988).

Building materials incorporating fly ash or slag generally have elevated ²²⁶Ra contents. Since these by-products have a low emanation coefficient (see Table 5.4), their use does not always result in a higher exhalation rate. This is illustrated in Table 5.5 on the basis of results of measurements on comparable small concrete walls. Although the addition of fly ash or phosphate slag does increase the radium concentration (up to 70-fold in this study), the increase in the exhalation rate is limited to a factor of 2 to 3 compared with ordinary concrete.

Table 5.5. The influence of industrial by-products on the exhalation rate (van Dijk and de Jong, 1990)

Building material	²²⁶ Ra concentration (Bq kg ⁻¹)	Emanation coefficient	Exhalation rate of ²²² Rn (mBq m ⁻² s ⁻¹)
Concrete	9.5	0.26	0.61 ± 0.08
Concrete containing fly ash	13	0.21	0.67 ± 0.03
Concrete containing granular fly as	h 55	0.05	0.56 ± 0.06
Concrete containing phosphate slag	710	0.01	1.72 ± 0.06

5.3 CONCENTRATIONS IN WATER

The concentrations in groundwater, surface water and drinking water are discussed in this section. The amount of Dutch measurement data in this field is extremely limited. Mattern (1973) analyzed 20 samples of groundwater taken for ²²²Rn in the province of Limburg. The concentrations ranged from about 500 to 20,000 Bq m⁻³ of water with an average of 4800 Bq m⁻³. Measurements of groundwater concentrations carried out in various other countries are summarized elsewhere (NCRP, 1988; Nazaroff et al., 1987; UNSCEAR, 1988). Surface waters have much lower ²²²Rn concentrations than groundwaters.

According to UNSCEAR (1988), they are expected to be similar to those of ²²⁶Ra, i.e. about 10 Bq m⁻³ (Hess et al., 1985; see also Table 2.2).

Both groundwater and surface water are used in the Netherlands for the production of drinking water. In most cases the unpurified water undergoes one or more purification steps during which a fraction of the ²²²Rn can escape. In a number of other cases the pumped-up water is transferred directly to the distribution system. The tap water obtained from groundwater in the above-mentioned study by Mattern showed a range of 100-20,000 Bq m⁻³. Since only a limited number of samples were analyzed, these data should be regarded as giving a rough indication only.

The ²²²Rn contained in tap water can be released during household use. The fraction released into a building depends on the manner in which the water is used, such as the degree to which it is heated, as shown in Table 5.6. The use-weighted mean transfer efficiency from water to air is 0.55. Using this value yields a mean of 0.65 x 10⁻⁴ for the ratio of the airborne ²²²Rn concentration to the concentration in water in a typical dwelling (Nazaroff et al., 1987). UNSCEAR (1988) has adopted a value of 1x10⁻⁴ for this ratio. Taking the UNSCEAR value and a ²²²Rn concentration of 20,000 Bq m⁻³ in drinking water, the contribution to the indoor airborne ²²²Rn concentration from water use is estimated to be about 2 Bq m⁻³ at the most. In general, however, this source contributes no more than about 0.6% to the ²²²Rn in the air of Dutch homes (see Table 2.3).

Table 5.6. Transfer efficiency of ²²²Rn from water to air, depending on use (Nazaroff et al., 1987)

Use	Fraction of 222Rn liberated	
	mean	
Dishwasher	0.95	
Shower	0.66	
Bath	0.42	
Toilet	0.30	
Laundry	0.92	
Drinking water and cleaning	0.34	
Use-weighted mean	0.55	

Data on the occurrence of 220 Rn in water are not available for the Netherlands. For a water-saturated soil, the 220 Rn concentration in the water, C_{water} , can be calculated using the equation:

$$C_{\text{water}} = A \rho \eta \epsilon^{-1}$$
 (5.1)

where A = the concentration of ²²⁴Ra in the soil (35 Bq kg⁻¹)

 ρ = the density of the soil (1600 kg m⁻³)

 η = the emanation coefficient (0.3)

 ϵ = the porosity of the soil (35%).

The equilibrium equivalent ²²⁰Rn concentration, which is calculated using the numerical values given in parentheses, is 48,000 Bq m⁻³. Because of the short half-life of ²²⁰Rn (56 s), its concentration in water, once it has been pumped up, will fall rapidly and not cause a significant increase in the indoor ²²⁰Rn concentration.

5.4 CONCENTRATIONS IN AIR

5.4.1 Indoor air

The ²²²Rn concentration in a building is subject to fairly wide variations, which can be attributed to a number of causes. One of the most important of these is broadly termed occupant behaviour (for example, with regard to window- and door-opening, heating and the like). As a result, the ²²²Rn concentration can vary by a factor of 3 to 4 within a few hours. In addition to these fairly rapid fluctuations there is also a day-to-day variation, caused by metereological conditions (e.g., wind speed and direction, water table and atmospheric pressure differences). These effects are shown in Figure 5.1, as well a third variation, namely, that from season to season.

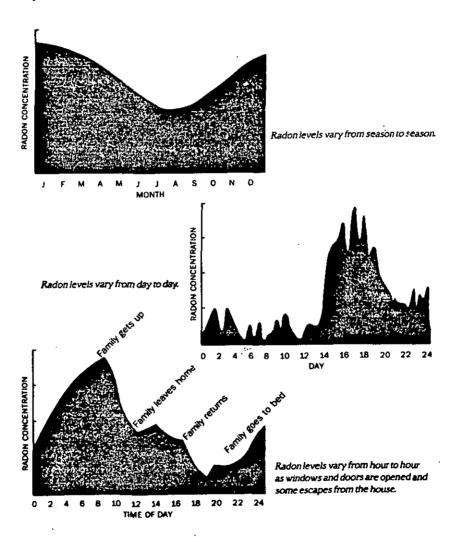


Figure 5.1. Variation of the ²²²Rn concentration with time; from top to bottom: the seasonal influence, the day-to-day variation, and the variation with time of day (NRPB, 1990).

The assessment of the radiation risk resulting from exposure to ²²²Rn is generally based on the annual average concentration. In the period 1982-1984, an investigation of this concentration in the Dutch home was conducted by the Kernfysisch Versneller Instituut (KVI) as part of the SAWORA research programme (de Meijer et al., 1986). The study covered a total of about 900 dwellings. The annual average ²²²Rn concentrations in living

rooms varied from 8 to 140 Bq m⁻³, with mean and median values of 29 Bq m⁻³ and 24 Bq m⁻³, respectively. The frequency distribution of the annual average concentrations is shown in Figure 5.2.

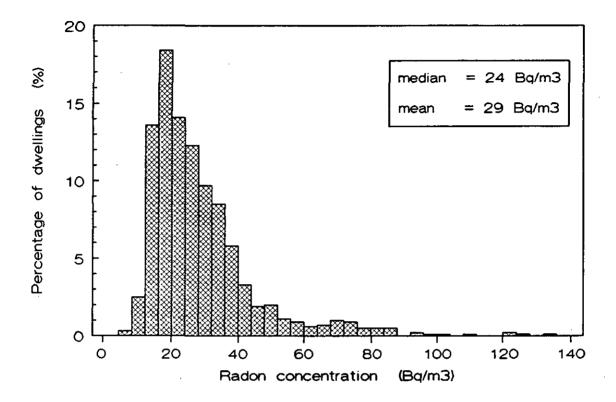


Figure 5.2. Frequency distribution of the annual average ²²²Rn concentrations in living rooms in the Netherlands.

Surveys of the type described here have also been conducted in many other countries (CEC, 1987; ICRP, 1987; UNSCEAR, 1988). The results from a number of these studies are presented in Table 5.7. The median ²²²Rn concentrations do not vary much, the approximate range being 20-60 Bq m⁻³. The same is true of the mean concentrations. The mean ²²²Rn concentration of 29 Bq m⁻³ in the Netherlands falls within the range of about 20-90 Bq m⁻³ observed in other European countries. However, the Netherlands does stand out in another respect. The distribution, presented in Figure 5.2, was found to have a much shorter tail than has been observed elsewhere. This implies that the number of houses with higher concentrations is markedly lower in the Netherlands than in the neighbouring countries. Taking a concentration of 100 Bq m⁻³ as an example, this percentage can easily be of the order of 5-10%, as against 0.7% in the Netherlands ICRP, 1987; de Meijer et al., 1986).

Table 5.7. Average 222Rn concentration obtained in indoor surveys (UNSCEAR, 1988)

Country	Median concentration Bq m ³	Mean concentration Bq m ³	σ _g (*)
Netherlands	24	29	1.6
Belgium	41	nr	1.6
Western Germany	40	49	1.8
United Kingdom	17	20 (**)	2.4
France	44	77	nr
Denmark	50	nr	nr
Norway	nr	90	nr
Sweden	46	85	nr
Finland	64	90	3.1
USA	35	61	2.8

(*) Geometric standard deviation

The buildup of the ²²²Rn daughter activity concentration in indoor air is determined by a combination of factors. An important parameter is the deposition rate of ²²²Rn daughters on surfaces such as walls. The ratio of the equilibrium equivalent radon concentration (EEC, see 1.2.2) to the radon concentration is the equilibrium factor F (equal to 1 if radon and all its decay products are in radioactive equilibrium). An average equilibrium factor of about 0.4 can be calculated from the data published by de Meijer et al., (1986). This value is in agreement with the average value calculated by UNSCEAR (1988) from 10 different indoor surveys. The values obtained in these studies lie in the range of 0.3-0.8. An average EEC of 12 Bq m⁻³ was found in the Netherlands (de Meijer et al., 1986).

The ²²²Rn concentration in a house is determined primarily by the source strengths. The soil beneath the house and the building materials used constitute the principal sources. Natural gas and domestic water supplies are minor sources of indoor ²²²Rn. In normal situations, the two together are estimated to contribute less than 1% to the ²²²Rn concentration in the indoor atmosphere (see Table 2.3). Radiation protection policy makes a distinction between ²²²Rn originating from natural gas or drinking water and ²²²Rn originating from soil or building materials. A risk limit of 10⁻⁶ a⁻¹/10⁻⁸ a⁻¹ has been proposed for the first two sources (VROM, 1991). This limit does not apply to the other two, but the policy aim is risk reduction.

Calculations using the model indicate that for a typical Dutch house the ratio of the ²²²Rn contribution from the soil to that from building materials is approximately 2 to 1. The results from the SAWORA survey showed no relationship between the soil type and the ²²²Rn concentration in the living room (De Meijer et al., 1986). It is known, however, that up to 1985 harbour sludge was one of the materials used in levelling up building land. In view of the composition of certain types of harbour sludge, Köster (1991) does not rule out the possibility that the use of these sludges could give rise to elevated concentrations inside dwellings.

^(**) NRPB, 1990

nr not reported

In addition to the sources, other factors also affect the indoor radon concentration. Measurements in houses show that the above guidelines give only an indication of the problem of indoor radon. The following factors are discussed below:

- 1. Thermal insulation
- 2. Concentrations in rooms other than the living room
- 3. Type of dwelling.

The study by the KVI revealed that the average ²²²Rn concentration is higher in houses where insulation measures, such as double glazing and wall insulation, had been taken. Higher concentrations probably do not result from these measures in themselves, but from the associated weather stripping, caulking around windows and plugging holes and openings in the building's shell, so greatly reducing the natural ventilation of the dwelling: For fixed source strengths, a decrease in the ventilation rate results in an increase in the ²²²Rn concentration.

Because the principal source of radon in a building is ordinarily the soil underneath it, it might be expected that rooms on a higher floor show a lower concentration. In general, this is indeed the case, although exceptions have been reported (van den Berg, 1990). In addition to the concentration in the living room, the KVI study also measured the concentration in another room on a higher floor, usually a bedroom. The ²²²Rn concentration was on average about 80% of that in the living room. The fact that bedrooms are aired for longer periods possibly also plays some role in the observed lower levels. A presumably very important parameter in the indoor radon issue is the type of dwelling. Only a limited number of studies have been made of the influence of this parameter on indoor concentrations. In the survey conducted in the Netherlands it was found that detached houses had the highest concentrations (de Meijer et al., 1986). The same phenomenon has been noted in Germany, where it was also found that radon concentrations were, in general, greater in multi-flat houses than in high-rise flats (UNSCEAR, 1988).

Because of the short half-life of ²²⁰Rn and consequently its short diffusion length, only the ²²⁰Rn produced in the surface layer of the building materials and the ground can escape into the air. Small barriers, for example in the form of a finishing coat or a fairly tightly sealed floor slab, already have a large effect on the amounts released and the resulting airborne concentrations. The ICRP (1987) gives a summary of the (little) literature on this subject. The average equilibrium equivalent concentration (EEC) of ²²⁰Rn lies in the range of 0.1-1 Bq m⁻³, which is mainly attributable to ²¹²Pb. This range agrees well with the limited Dutch measurement data on ²²⁰Rn. The (instantaneous) EEC_{Rn222} was measured in six dwellings as part of the SAWORA research programme. The observations ranged from 0.1 to 0.7 Bq m⁻³, with a mean of 0.4 Bq m⁻³ and a standard deviation of 0.2 Bq m⁻³ (Hogeweg, 1986). In this integrated criteria document, the value used for the EEC_{Rn220} in the further calculations is 0.5 Bq m⁻³ (ICRP, 1987; UNSCEAR, 1988).

As far as we know, no radon measurements have been performed in the Netherlands in buildings other than dwellings. Examples of such buildings are schools, factories or offices. It is assumed in this report that the average working-day concentrations do not differ from those in houses (living rooms).

Given the concentrations in soil gas (see 5.1) and groundwater (see 5.3), relatively high

concentrations can be expected in caves and thermal baths, respectively. This is also supported by foreign data (Wilkening, 1990). The concentrations in tunnels will be much lower than in caves because of the use of forced ventilation, but here, too, measurement data are not available.

5.4.2 Outdoor air

In addition to the measurements of ²²²Rn levels in indoor air, the annual average concentration in outdoor air was determined at 250 locations in the Netherlands, and found to range from 1 to 10 Bq m⁻³ (De Meijer et al., 1986). The frequency distribution is shown in Figure 5.3. The median and mean concentrations are 3.0 Bq m⁻³ and 3.3 Bq m⁻³, respectively. These are normal values for a coastal region (see 3.1.1). The contribution of outdoor air to the average indoor concentration is therefore about 10%. The annual average concentration is fairly constant. Measurements made over a period of four years indicated that at a given location, the annual variation is less than 30% (Put and de Meijer, 1988).

Figure 5.4 shows the annual average outdoor concentration in the Netherlands (de Meijer, 1991). The highest concentrations are observed west of Delfzijl. Other areas with much higher-than-average concentrations are Zealand, Flanders, South Limburg, parts of the Betuwe and Salland, the Haarlemmermeer Polder, and the Groningen-Frisian clay region. Apparently the exhalation rate from soil is higher at these locations than elsewhere.

The ²²⁰Rn concentrations are of the same order as those of ²²²Rn. The ICRP (1987) gives a range of 0.5-20 Bq m⁻³ in the air layer 1-10 m above ground level. The equilibrium-equivalent concentration of ²²⁰Rn is much less than that that of ²²²Rn, which is due to the large difference in the equilibrium factor. A value of 0.8 has been adopted for the equilibrium factor for ²²²Rn in outdoor air (UNSCEAR, 1988), and of 0.04 for ²²⁰Rn (ICRP, 1987). This parameter has not been measured in the Netherlands.

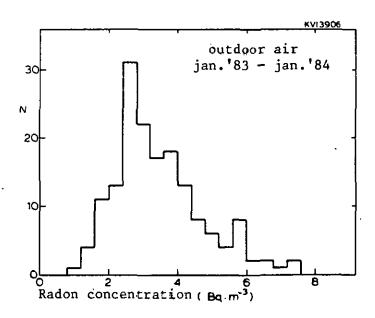


Figure 5.3. Frequency distribution of the annual average ²²²Rn concentration in outdoor air.

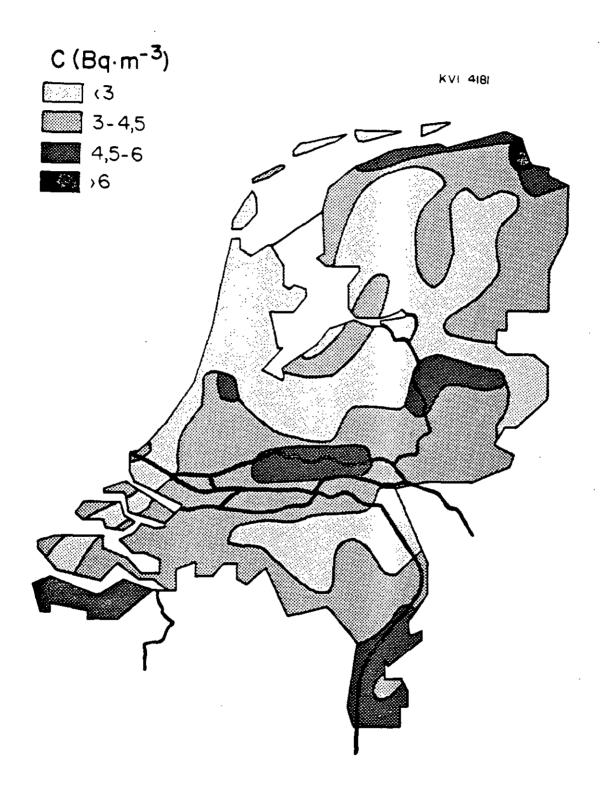


Figure 5.4. Annual average ²²²Rn concentration in outdoor air in the Netherlands.

5.5 EXPOSURE LEVELS

Exposure of humans to radon daughters occurs mainly through inhalation (see 6.1.3). Three components of the radon daughter exposure have to be distinguished:

- indoor exposure at home
- indoor exposure elsewhere (e.g. office, school)
- exposure out of doors

UNSCEAR (1982) assumes a mean occupancy factor of 0.8 for residence indoors, without distinguishing between residence at home and elsewhere. Policy also makes no distinction between houses and other buildings (VROM, 1991). In the United Kingdom, however, a survey on the occupancy factor in private houses, other buildings, and out-of-doors as a function of time of day was carried out (Brown, 1983). On the basis of these data, the ICRP (1987) has determined corrected mean occupancy factors. It estimated the annual average radon daughter exposure using these factors, taking into account the diurnal variation of the radon concentrations indoors and outdoors. The occupancy factors are included in Table 5.8.

The table also summarizes the annual average radon concentrations in the Netherlands. Measurement data on concentrations in factories, schools and office buildings are not available. It is assumed that they are the same as the mean concentrations in living rooms. The mean annual exposure to radon daughters (PAEE) has been estimated using these data and the conversion factors given in 1.2.2.

On the basis of the survey of ²²²Rn concentrations in Dutch living rooms, it can be calculated that the relative standard deviations of the mean EECs given in Table 5.8 are 4-5%. The relative error in the PAEE is therefore of the same magnitude.

Table 5.8. Occupancy factors, mean concentrations, equilibrium equivalent concentrations and annual potential alpha energy exposures

Residence	Occupancy	²²² F	₹n	<u> </u>	²²⁰ Rn		
area factor	(Bq m ⁻³)	(Bq m ⁻³)	PAEE (μJ m ⁻³ h)	EEC (Bq m ⁻³) (μJ	PAEE m ⁻³ h)		
At home:							
living room	0.37	29	12	208	0.5	123	
bedroom	0.33	23	9	147	0.5	109	
Indoors							
elsewhere(*)	0.18	29	12	101	0.5	60	
Outdoors	0.12	3	2	12	0.2	16	
Total (rounded off)	1.00			470		310	

^{*)} Estimated concentrations

EEC Equilibrium equivalent concentration

PAEE Potential alpha energy exposure

5.6 SUMMARY AND CONCLUSIONS

A member of the Dutch population receives the greatest ²²²Rn exposure in his or her home. The median indoor concentration in the Netherlands is 24 Bq m⁻³, which is within the range of 20-60 Bq m⁻³ observed in the neighbouring countries. The mean ²²²Rn concentration in Dutch living rooms (29 Bq m⁻³) falls within the 20-90 Bq m⁻³ range measured in other European countries. However, the variability around the mean is in most of these countries markedly greater than in the Netherlands, thus implying that the percentage of dwellings with high concentrations is relatively higher there than in the Netherlands.

A second source of the ²²²Rn exposure is formed by buildings other than private homes. Examples include schools, factories and offices. Data on the radon concentrations in these buildings are not available in the Netherlands. Policy makes no distinction as to type of building, but considering the estimated 20% contribution to the exposure from residence indoors, excluding homes (see Table 5.8), research into this component of the radon exposure seems desirable.

The principal sources of indoor ²²²Rn are the soil beneath the house and the building materials incorporated in its structure. The ratio of the contribution from the soil to that from building materials is approximately 2:1. Natural gas and water usage contribute about 1% to the mean concentration. The contribution of outdoor air is about 10% on average.

The exhalation rates from the building materials used in the Netherlands are relatively well known. Future research on exhalation rates can therefore be limited to new building materials, for example, incorporating a certain number of industrial by-products. The soil generally appears to be the principal source of ²²²Rn in the Netherlands, certainly in dwellings with elevated ²²²Rn concentrations. Given the importance of the soil and the fact that the exhalation rate has been determined at 10 locations only, this is a considerable gap in our knowledge. In addition, little attention has as yet been paid to ²²⁰Rn.

6 EFFECTS

In principle, radon can enter the body by three routes; namely, through the skin, by ingestion and by inhalation. Inhalation is by far the most important route of entry of radon into animals or humans. Outdoors, humans and animals are exposed to radon, originating primarily from the soil (see Chapter 2). The soil is ordinarily also the principal source of radon in dwellings, entering via crawl spaces and cellars and the ventilation system. In addition, radon exhalation from building materials contributes to indoor concentrations: People are also exposed to gamma radiation, arising from short-lived daughter products of radon contained in soil and building materials. The latter aspect is briefly discussed in Chapter 7.

Epidemiological studies of underground miners constitute the main basis for the evaluation of the biological effects of radon in humans. Death from lung cancer is generally considered to be the most important effect of exposure to radon. Recently, however, other forms of cancer (leukaemia and mammary tumours) have also been linked to enhanced radon exposure. Nevertheless, the emphasis in this chapter will be on lung cancer mortality. To extrapolate the data from studies on miners to the general population, it is essential to know the influence of a number of dosimetric factors, such as aerosol size, quality factor for alpha radiation, metabolism of radon (daughters) and dosimetric models. These models are also needed to enable estimation of the organ doses from inhaled ²²⁰Rn and its daughters.

Before considering the effects of radon exposure on humans, a number of dosimetric factors are discussed, as well as data from animal studies on effects from ²²²Rn. An evaluation of interaction effects between radon and other agents completes the section on human toxicity. The chapter concludes with paragraphs on ecotoxicity and toxicity to livestock.

Exposures to radon will be expressed as much as possible in the unit Bq m⁻³ a (one year of continuous exposure to 1 Bq m⁻³ (EEC), and/or radon gas concentration; see 1.2.2 for the definition of EEC). However, the WLM (see 1.3.1) will also be used as a unit of exposure. The WLM is a historically established term and is used in virtually all publications in the field of radon exposures. For the sake of consistency with the literature, the WLM will be used in this integrated criteria document, although it is not an SI unit.

6.1 HUMAN TOXICITY

6.1.1 Aerosols

The radiation dose resulting from inhalation of radon and radon daughters is determined not only by the quantity of radioactivity inhaled, but also by the aerosols present in the inhaled air.

Aerosols are microscopic solid or liquid particles suspended in air (for example, dust or minute water droplets). The importance of these aerosols lies in the fact that the daughters of radon, unlike radon itself, are solids at room temperature. When radon gas decays in the atmosphere, the daughters are formed as electrically charged atoms, which are originally also in the gas phase, but which, owing to their solid nature and electric charge,

have a strong tendency to attach themselves to other solids or liquids (as in condensation of water vapour). This deposition may take place on aerosol particles. It is a process governed by chance, so that in practice not all the daughter atoms will become attached (attached fraction). The remaining free daughter atoms are referred to as "free fraction" or "unattached fraction", f.

The attached and unattached fractions behave differently in the respiratory tract. Because the unattached fraction has a high deposition efficiency and is very mobile, all inhaled free daughter atoms are deposited in the upper airways (nasopharynx and large bronchi) and deliver their energy there. However, the behaviour of attached daughter atoms is determined by the behaviour of the aerosols, which are relatively heavy and consequently not very mobile. This enables the attached atoms to penetrate much deeper into the lung, or even to be exhaled again, depending on the aerosol particle size. Within the respiratory tract, the most radiosensitive tissue is considered to be the bronchial epithelium (ICRP, 1987; BEIR IV, 1988), so that it is important to know the location of the target cells in the lung; this depends greatly on the fraction of unattached daughters in the inhaled air, and on the size of the carrier aerosol of the attached daughter atoms. The activity median diameter (AMD) is used to describe the size of the aerosols to which radon daughters have attached themselves. It is implicitly assumed that the size distribution of these aerosols is log-normal.

In practice, the average values of both the unattached fraction and the aerosol size are used in characterizing the indoor atmosphere. For 222 Rn progeny, the ICRP (1987) gives typical values of 3% (range 1-5%) for the unattached fraction and of 0.15 μ m (range 0.1-0.3 μ m) for the AMD. The values reported by Nazaroff and Nero (1988) for 222 Rn progeny are 5% (range 2-15%) for the unattached fraction and 0.1 μ m (range 0.01-0.2 μ m) for the AMD. For 220 Rn progeny, Nazaroff and Nero (1988) employ an AMD of 0.2-0.3 μ m, and assume an unattached fraction of 2% for the 212 Pb daughter. They consider the contributions of the other 220 Rn daughters negligible for dose calculation. Jacobi and Eisfeld (1980), however, include all daughter nuclides in the dose calculations for 220 Rn, and assume an unattached fraction of 100% for the 216 Po daughter and 0% for the other daughters. They use a value of 0.25 μ m for the AMD.

6.1.2 The quality factor for alpha particles in radiological protection

Equal doses of different types of radiation do not (necessarily) produce the same degree of biological effect. This is because the biological effect depends on, for example, the microscopic distribution of energy dissipation (radiation quality). The ICRU has introduced a number of quantities for quantifying the radiation quality, including linear energy transfer, LET, lineal energy, y, and relative biological effectiveness, RBE (ICRU 33, 1980).

On the basis of general observations and theoretical considerations in radiobiology, the ICRU 40 (1986) has related the quality factor, Q, and the lineal energy, y, determined in a 1 μ m diameter sphere of ICRU tissue (muscle). Since any irradiation entails a range of values of lineal energy, it is often necessary to define an effective quality factor, \overline{Q} :

$$\overline{Q} = \frac{1}{D}D(y) \cdot Q(y) \cdot d(y)$$
(6.1)

where d(y) is the distribution of absorbed dose in y. The values of Q(y) have been so scaled that the effective quality factor, \overline{Q} , is 1.0 for photons of near 100 keV.

For alpha-emitting radionuclides in ICRU 40 (1986) an effective quality factor for the entire range of alpha particles of about 25 is derived (see Figure 6.1). This value differs from that of the ICRP (1991) which gives a radiation weighting factor, W_R , of 20 for alpha particles, based on a relation between the quality factors and the linear energy transfer. Thus different values for the quality factor are recommended for calculating the dose equivalent. There is no international agreement on the value to be adopted for the quality factor.

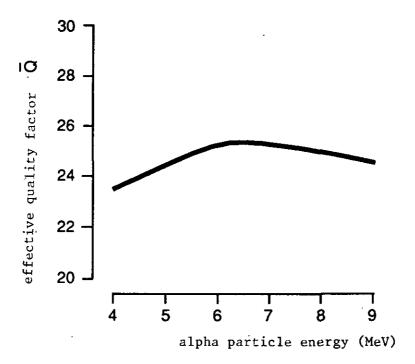


Figure 6.1. Calculated values of the effective quality factor, \overline{Q} , for the entire range of alpha particles of a given initial energy.

6.1.3 Metabolism and organ doses

In principle, radon can enter the body by three routes, namely, through the skin, by ingestion and by inhalation. The first possibility can be ignored because absorption through the skin is negligible compared with intake by inhalation (Jacobi and Eisfeld, 1980). Ingestion plays a role when drinking radon-rich water, and the stomach has been identified as the organ receiving the largest dose. However, estimates indicate that the lung dose resulting from inhalation of radon released during domestic water use is one order of magnitude greater than the stomach dose due to ingestion (Nazaroff and Nero, 1988). Since radon originating from other sources also enters the body by inhalation, this exposure route contributes by far the most to the detrimental effects of radon.

After inhalation, radon is absorbed into the blood from where it is distributed throughout the whole body. Radon dissolves to a greater or lesser extent in body fluids and organs. The resulting radiation dose can be estimated using a mathematical model. Only the dose from alpha radiation is important because not only are alpha particles more energetic than

beta particles and gamma rays but their biological effects are greater. The quality factor assigned to alpha particles is about 20. Table 6.1 shows the dose rates to various organs from the inhalation of pure radon, calculated with the Jacobi-Eisfeld model (Jacobi and Eisfeld, 1980). This model takes account of the difference in radon solubility in the various organs, build-up of radon daughter activities, retention and translocation, and physical and biological half-lives. The breathing rate (m³ h¹) is not important here because we are concerned with pure radon gas; it is this concentration in the inhaled air which determines the equilibrium concentration in the body.

Table 6.1. Contribution of various organs/tissues to the effective dose equivalent rate, $\dot{H}_{\rm eff}$, from radon alone at continuous exposure to 1 Bq m³ of 222 Rn or 220 Rn in the inhaled air (Jacobi and Eisfeld, 1980). The breathing rate does not play a role here (see text).

Organ/tissue	Contribution to H _{eff} ()	uSv a ⁻¹ per Bq m ⁻³ of radon gas) 200Rn
Lung	0.79	0.62
Liver	0.05	0.03
Kidney	0.06	0.12
Spleen	0.05	0.01
Red bone marrow	0.10	0.04
Bone surfaces	0.02	0.11
Other tissues	0.45	0.02
Total	1.52	0.95

Radon daughters, unlike radon itself, are not chemically inert. Moreover, because they are solids at room temperature, they can deposit in the respiratory tract when inhaled. They may be absorbed from the lung into the blood, or be cleared by mucociliary action to the throat for swallowing. After swallowing the radon daughters enter the gastrointestinal tract, from where they may be transferred to blood and carried to other organs of the body. They can be taken up more or less selectively by various organs. For example, lead is a bone seeker while the main target organ of bismuth is the kidneys (ICRP 30, 1979). The associated radiation dose can be estimated using a mathematical model, for which, as previously mentioned, only the alpha radiation is of dosimetric significance. The results obtained with the Jacobi-Eisfeld model are given in Table 6.2. It can be seen from the tables that of all organs, the contribution to the effective dose equivalent from the lungs is by far the most important. Other model calculations have confirmed this observation: Nazaroff and Nero (1988) noted that the doses to organs other than the respiratory tract due to 222Rn are about two orders of magnitude lower than the dose to the lung. For ²²⁰Rn they give values which, except for the lung dose, are a factor of 4 to 40 lower than those in Table 6.2.

Contributions of various organs to the effective dose equivalent rate, $\dot{H}_{\rm eff}$, from radon daughters alone at continuous exposure to 1 Bq m³ of ²²²Rn or ²³⁰Rn in the inhaled air, using an equilibrium factor, F, of 0.4 and 0.025, respectively. A breathing rate of 1.2 m³ h⁻¹ has been assumed. The corresponding effective dose equivalents per WLM exposure (Jacobi and Eisfeld, 1980) are given in parentheses (mSv WLM⁻¹). For the definition of the unit WLM, see 1.3.2.

Organ/tissue	Contribution t	o Ĥ _{eff} (μSv	a ⁻¹ per Bq	m ⁻³ of radon gas)
Lung	44.8	(8)	14.4	(3)
Liver	<0.5	(<0.01)	0,34	(0.07)
Kidney	0.22	(0.04)	1.78	(0.37)
Spleen	<0.05	(<0.01)	0.05	(0.01)
Red bone marrow	<0.05	(<0.01)	0.34	(0.07)
Bone surfaces	<0.05	(<0.01)	1.15	(0.24)
Other tissues	<0.05	(<0.01)	<0.05	(<0.01)
Total	45	(8)	18	(4)

Although it is difficult to draw quantitative conclusions from such estimates about doses to organs other than the lung, it can be concluded qualitatively that these doses are small compared with the lung dose. It is also seen in the tables that the lung dose from radon itself, except for very extreme conditions (exceptionally low equilibrium factor, F), is much smaller than the dose from the daughter products. Therefore, only the last dose is usually taken into account (ICRP 50, 1987; BEIR IV, 1988).

Comparison of the total doses from ²²²Rn daughters with those from ²²⁰Rn daughters shows that the contribution per unit of exposure from ²²⁰Rn daughters is a factor of 2 to 3 lower than that from ²²²Rn daughters. Combining this with the fact that in most situations air contains less ²²⁰Rn than ²²²Rn, it may be concluded that the most significant radiation dose from radon inhalation is that from inhalation of ²²²Rn.

The Jacobi-Eisfeld model estimates a dose equivalent rate to body fat of $10~\mu Sv~a^{-1}$ per Bq m⁻³ at continuous exposure to ²²²Rn alone. Because adipose tissue has not been included in the ICRP system as a critical organ for the calculation of the effective dose equivalent (ICRP, 1991), it is also absent from Table 6.1, despite the fairly high dose equivalent predicted by the model. This prediction arises from the fact that ²²²Rn is much more soluble in fat than in an aqueous environment (see Table 1.1). In a recent publication (Henshaw et al., 1990) it was argued that, given these high fat doses and the fact that red bone marrow contains fat cells, the dose to bone marrow from ²²²Rn could be much higher than previously assumed. Henshaw et al. calculated an annual dose equivalent of 5 μ Sv per Bq m⁻³ of ²²²Rn, corresponding to a contribution of 0.6 μ Sv per Bq m⁻³ to the annual effective dose equivalent. This is six times higher than the value in Table 6.1. Henshaw et al. believe that a considerable percentage of leukaemia cases may be attributed to ²²²Rn, and in the same article demonstrated a statistically significant correlation between leukaemia incidence and ²²²Rn exposure in the home.

A similar situation could apply to the mammary gland; as it also contains fat, it could give rise to fairly considerable radiation doses from ²²²Rn. The mammary gland is one of the organs most susceptible to radiation carcinogenesis, which could give rise to the

supposition that exposures to high ²²²Rn levels could also induce breast cancer. Indications along these lines can be found in an (unpublished) animal experiment in France, which included a pilot study attempting to induce lung tumours in female rats by ²²²Rn inhalation. However, instead of developing lung tumours, the animals died prematurely from mammary tumours. Researchers at the ITRO-TNO exposed female rats to ²²²Rn. Measurements on adipose tissue and mammary gland tissue showed that the activity concentrations in these tissues were similar to that in lung tissue.

Consideration of the above findings suggests that ²²²Rn can cause leukaemia and breast cancer. Instead of taking only the lung as a critical organ for cancer induction, bone marrow and mammary tissue should also be regarded as such, and further research into the risk of leukaemia and mammary tumour induction by ²²²Rn is needed.

6.1.4 Lung dosimetry

In the dosimetry of radon (daughters) in the human lung, an attempt is made to derive a dose conversion coefficient linking exposure to radon and radon daughters in air (in Bq m⁻³ or WL) to the dose equivalent in lung tissue (hereafter referred to in short as lung dose). The purpose of this is two-fold: firstly, using the estimated lung dose (and, if desired, the doses in other organs), the effective dose equivalent can be calculated. From this value the ultimate mortality risk can be obtained via the risk coefficient of 5x10⁻² Sv⁻¹ (ICRP, 1991). Secondly, for a given exposure level (in Bq m⁻³ or WL), lung doses can be estimated under exposure conditions to be found in a mine or a home. From these lung doses, a conversion factor K can be derived. By using this factor, it is possible to extrapolate the risk value for miners, estimated from epidemiological data, to members of the general public.

Lung models

The lung dosimetry of radon daughters is based primarily on calculations using models of the human airways. In each model, the respiratory tract is divided into three regions: the naso-pharyngeal (N-P), the tracheo-bronchial (T-B), and the alveolar-interstitial or pulmonary (P) regions. Since most lung tumours arise in the bronchial airways, many models focus on the T-B region and, to a lesser extent, on the P region.

The basis of a mathematical model for the T-B region is a geometrical model of the bronchial tree, which describes the branching and dimensions of the bronchi. Each branching yields a class of bronchi, also called a generation, which differs from preceding generations as to dimensions (but also physiologically and histologically). Commonly used geometrical models are: 1) the so-called Weibel "A" model, with a symmetrical branching structure with 16 generations, and dimensions which are represented by formulae in terms of the generation number (based on measurements in human lungs), and 2) the so-called "Y-S" model, based on a cast of a human lung.

Because the bronchial generations have different properties, they must be considered separately in the mathematical model. Such a separate part of the model is also called a compartment. The various compartments may be linked through biological or physiological transport processes, each assumed to be governed by first-order kinetics. This means that all particles (here: radioactive atoms) of a given type have the same probability per unit time of being transported from one compartment to another. The transport process can be described by a characteristic half-time T. Radioactive decay is a good example of a process of first-order kinetics, but for many biological processes it is

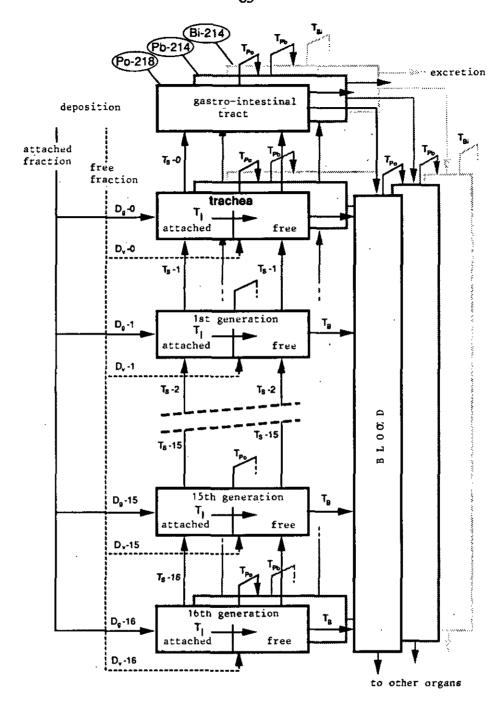
not known whether they answer to these kinetics. However, the uncertainties resulting from this may be no greater than those resulting from inaccuracies in physiological data (BEIR IV, 1988).

Daughter atoms attached to aerosols behave differently from free atoms; the deposition probabilities in each generation of the bronchial tree are different and, after deposition, attached daughter atoms must first desorb from the carrier aerosols before they can be absorbed into the blood. Therefore, each bronchial generation is divided into two compartments, one for the attached atoms and one for the free atoms deposited there. Moreover, such a model must, in principle, be constructed for each of the four daughter nuclides separately, which then interconnect through the process of radioactive decay. Upon decay of a parent nuclide to a daughter nuclide, the atom concerned will move from a particular compartment in the parent part to the corresponding compartment in the daughter part. As an example, the compartmental model used in the Jacobi-Eisfeld model is shown in Figure 6.2; similar compartmental models are used for the N-P and P regions.

Finally, the compartmental models make it possible to calculate how much activity is present, from which nuclide and in which tissue it is as a function of time. To calculate the doses in the various tissues from this, it is necessary to analyze microscopically the radiation energy which comes free due to a given activity. This is because the major part of the radiation energy is emitted in the form of alpha particles, which in tissue have a range of only a few tens of micrometres, corresponding to a few cell layers. Moreover, the largest energy deposition occurs at the end of the track of an alpha particle, so that it is very important to know the depth of the most radiosensitive cells below the bronchial surface.

In addition to the geometric and the compartmental models outlined above, every lung model contains a large number of physical and biological parameters to which a value must be assigned. The values used in the different models can vary greatly, because each model assumes different exposure conditions (variables), and because the constants are not all equally well-known. The parameters and values given below are those used in practice:

- deposition probability of aerosols after inhalation; this is highly dependent on the bronchial generation and a function of the breathing rate.
- radioactive decay constants for the different nuclides; these are very well-known.
- energy deposition along the track of an alpha particle; is well-known.
- diffusion coefficient of unattached atoms in air (0.005-0.05 cm² s⁻¹).
- clustering of aerosols in the airways; yes or no.
- equilibrium factor, F, of the 222Rn(-daughter) mixture in the inhaled air; values of 0.4-0.6 are usually used for domestic environments.
- the fraction unattached, f; about 1 to 15% (average 3 to 5%).
- AMD and aerosol size distribution. The AMDs used range from 0.05 to 0.3 μm for domestic environments. Some model calculations assume aerosols the size of the AMD value, while others take account of the size distribution of the aerosols around this AMD.
- type of aerosol; determines the rate of desorption of attached atoms (see also below).
- rate of mucus transport in the bronchial tree; this is dependent on the bronchial generation and remains uncertain: 1-2 cm min⁻¹ for the trachea to a few mm h⁻¹ for the terminal bronchioles.
- rate of absorption from the bronchial epithelium into the blood (half-time T_B between 4 and 40 min).



Compartments for the T-B region as used in the Jacobi-Eisfeld model. The arrows represent the transport processes. The deposition process can be characterized by the deposition probability D:

D_s-n: deposition probability for attached atoms in nth generation (n=0-16)

D_f-n: deposition probability for free atoms in nth generation. The other transport processes can be characterized by the half-time T associated with each process:

T_B: half-time for absorption from bronchi into blood

T_D: half-time for attached atoms to desorb from carrier aerosol

T_M-n: half-time for mucus transport in nth generation (n=0-

16) $T_{\rm bh}/T_{\rm bh}/T_{\rm hi}$: half-time for radioactive decay of polonium, lead

 $T_{Po}/T_{Pb}/T_{Bi}$: half-time for radioactive decay of polonium, lead and bismuth respectively.

- thickness of the mucous layer of the tracheobronchial epithelium; the thickness is dependent on the bronchial generation.
- radiation-sensitive cells in the bronchial epithelium; basal cells, mucus cells and/or cells in the pulmonary region are sometimes considered sensitive targets.
- rate of desorption, i.e. the rate at which attached atoms are detached from the carrier aerosol after deposition; depends on the type of aerosol and can vary from a few minutes to 10-20 hours; it is taken to be 5-10 hours for aerosols in indoor air.
- breathing through the nose or mouth; a fraction of the inhaled activity can be lost by filtration in the nose. The effect is a lower lung dose; the nasal cavity is not considered to be at risk.
- breathing rate; is highly dependent on the level of activity of a person; values of 0.5 to 2 m³ h¹ are used (1.2 m³ h¹ for "light" activity). The breathing rate influences the deposition probability of aerosols and free atoms.
- tidal volume; also depends on the level of activity of a person. A value of 0.5 to 1.5 litres per breath is assumed ("normal": 1 litre). This value, like the breathing rate, influences the deposition probability.

The different lung models do not always take into consideration all the parameters mentioned. For example, some models ignore uptake of nuclides into the bloodstream, while others disregard the clustering of aerosols in the airways. From another point of view, there are also factors which in reality do play a part but which are not included in any of the models because too little is known about them, for example, the non-uniform deposition at bronchi bifurcations.

Tissue doses and conversion coefficients

The estimated dose to the bronchial tissue in the various models can vary widely for the same exposure level, due to the uncertainty and possible variation in the values of many of the parameters discussed above, and differences in the loci of the bronchi for which the dose was calculated. The BEIR IV (1988) reports that the highest and lowest estimates of the doses per unit exposure differ by more than two orders of magnitude. However, if a linear dose-effect relationship without threshold is assumed for cancer induction, and if the radiosensitivity of the bronchial epithelium is the same over the whole bronchial region, only the average dose to the bronchial epithelium is important for the risk of a bronchial cancer. The differences in the mean bronchial dose per unit exposure are much smaller for the various models. Moreover, if the variation due to other assumptions on the values of some variables (for example, AMD, unattached fraction) is ignored, the models differ only by a factor of 2 to 3. The BEIR IV report (1988) shows these differences for three principal lung models, namely, those of Harley and Pasternak (1982) (H-P model), Jacobi and Eisfeld (1980) (J-E model), and James et al. (1980) (J-B model), as a function of particle size (AMD), per cent unattached fraction, and breathing rate (Figure 6.3). While these models differ in several important respects, including clearance mechanisms, cells at risk and lung morphometry, the differences in the dose in the relevant region calculated by the three models are less than a factor of 2 for all conditions. With respect to the mean bronchial dose, the ICRP (1987) too has concluded that the results from the different dosimetric models for given exposure conditions agree to within a factor of 2.

In the determination of the dose conversion coefficients for 222 Rn, the ICRP assumed an unattached fraction of 3% and an AMD of 0.15 μ m. A separate conversion coefficient is given for indoors and outdoors, because the breathing rate indoors is on average somewhat lower (0.75 m³ h⁻¹ and 1 m³ h⁻¹, respectively).

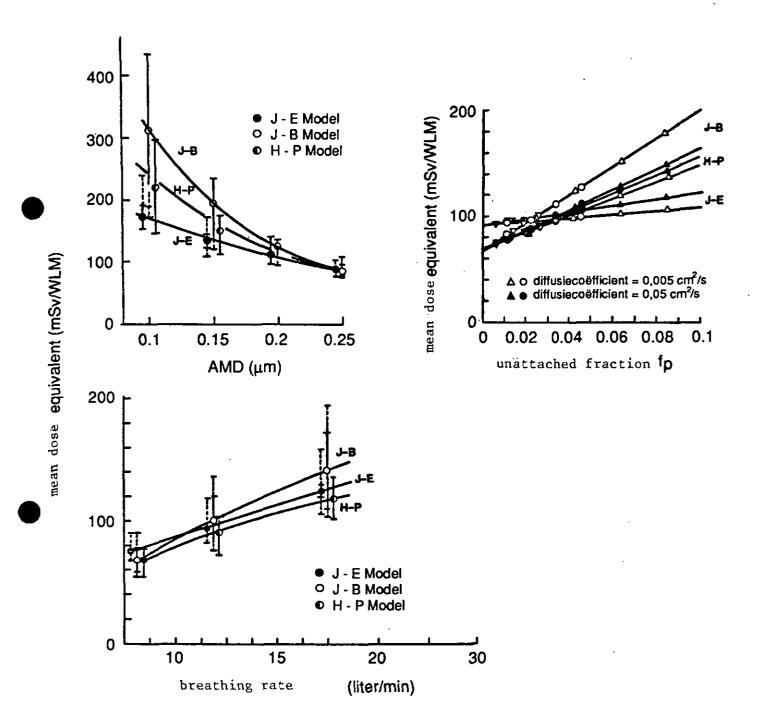


Figure 6.3. Mean bronchial dose equivalent per WLM exposure for 222 Rn, as a function of AMD, per cent unattached fraction, and breathing rate for three different dosimetric models. The differences are never greater than a factor of 2 for all conditions (BEIR IV, 1988).

Table 6.3. Conversion coefficients for the (effective) dose equivalent per unit exposure to ²²²Rn. To obtain the effective dose equivalent, a weighting factor of 0.06 has been assumed for both the bronchial epithelium and the pulmonary tissue. The last column gives the ratio, K, of the dose in homes to that in mines per unit of exposure.

Source	Dose equivale H (mSv a-1 p [(H (mSv WLM-1	er Bq m ⁻³ EEC)	
	bronchial	pulmonary	
ICRP indoors	1.3 (94)	0.18 (13)	
outdoors BEIR IV	1.8 (125)	0.24 (17)	
Nazaroff and Nero Jacobi and Eisfeld	3.5 (250) 1.5 (110)	0.31 (22)	

Source	Effective dose equivalent from ²²² Rn $\dot{H}_{\rm eff}$ (mSv a ⁻¹ per Bq m ⁻³ EEC) [$H_{\rm eff}$ (mSv WLM ⁻¹)] whole body	K (ratio of dose in homes to dose in mines)
ICRP indoors	0.09 (7)	0.8
outdoors	0.12 (9)	1.0
BEIR IV	• •	0.9
Nazaroff and Nero	0.21 (15)	1.3
Jacoby and Eisfeld		

(Values between brackets: doses in mSv WLM'1)

Table 6.4. Conversion coefficients for the (effective) dose equivalent per unit exposure to 220Rn. To obtain the effective dose equivalent, a weighting factor of 0.06 has been assumed for both the bronchial epithelium and the pulmonary tissue.

Source	Dose equivalen 20Rn H (mSv a per H (mSv WLM-1) bronchial		Effective dose equivalent from ²²⁰ Rn H _{eff} (mSv a ⁻¹ per Bq m ⁻³ EEC) H _{eff} (mSv WLM ⁻¹) whole body
ICRP Nazaroff and Nero Jacobi and Eisfeld	4.4 (23) 10 (50) 6 (30)	1.3 (7) 3 (15)	0.44 (2.3) 0.6 (3) 0.8 (4)

(Values between brackets: doses in mSv WML-1)

Also, a separate conversion factor K is given for indoors and outdoors to permit conversion of the doses to environmental conditions found in a mine. The various conversion coefficients for ²²²Rn are listed in Table 6.3 and those for ²²⁰Rn in Table 6.4. In its report, BEIR IV considers a K for ²²²Rn only and, moreover, gives no dose conversion coefficients. The BEIR IV model calculations are only used to estimate the conversion factor K. The value of 0.9 given in Table 6.3 can change (0.8-1.3) under other conditions (breathing, unattached fraction).

Nazaroff and Nero (1988) give conversion coefficients for ²²²Rn which are considerably higher than those reported by other investigators. This is due mainly to the fact that they assumed a much larger unattached fraction in homes than that assumed by, for example, the ICRP. They assumed a value of 2-15% for f, resulting in a bronchial epithelial dose of

120-500 mSv WLM⁻¹. The reported value of 250 mSv WLM⁻¹ is a "typical" value, which yields a conversion coefficient for the effective dose equivalent of 15 mSv WLM⁻¹. Nazaroff and Nero (1988) noted that this is substantially higher than the value of 5 mSv WLM⁻¹ adopted by UNSCEAR (1982), and therefore suggested a range of 5 to 15 mSv WLM⁻¹, with 10 mSv WLM⁻¹ as a reference value.

Finally, Jacobi and Eisfeld (1980) give dose conversion coefficients for 222 Rn as a function of the fraction unattached, f. An f of 5% has been assumed for the values in Table 6.3. In practice, it also appears possible to estimate the dose from domestic exposure to 222 Rn without knowing precisely the equilibrium factor F or the unattached fraction f (Nazaroff and Nero, 1988). This is because model calculations and experimental studies of the indoor atmosphere have shown that F generally increases and f decreases with aerosol concentration. Since for a given concentration of 222 Rn gas the dose generally increases with increasing F while it decreases with decreasing f, the two effects largely cancel each other out. Nazaroff and Nero recommend a conversion factor between radon gas concentration and bronchial epithelial dose of 50 μ Gy a⁻¹ per Bq m⁻³. Using a quality factor of 20 for alpha radiation and a tissue weighting factor of 0.06 for the bronchial epithelium, a conversion factor for the effective dose equivalent of 0.06 mSv a⁻¹ per Bq m⁻³ of 222 Rn gas can be calculated (assuming an equilibrium factor F of 0.4, this corresponds to 0.15 mSv a⁻¹ per Bq m⁻³ EEC). An uncertainty in this value is not given.

Age dependence

In principle, the dose from a given exposure is age-dependent because the dimensions as well as the breathing rate and tidal volume change with age; this may affect the dose per unit exposure. However, the effects are small: Nazaroff and Nero (1988) show that in children under 10 years, the dose to the larger bronchi is 1 to 1.2 times as much and that to the bronchioles up to twice as high as in adults.

ICRP Publication 50 (1987) mentions that for children, the dose models indicate a bronchial dose per unit exposure of twice that in adults. BEIR IV (1988), too, notes that for children the dose may be somewhat higher. The OECD/NEA (NEA, 1983) has recommended that age dependency of effective dose equivalent for exposure of the whole population be neglected.

Conclusions

Given the small difference between the dose conversion coefficients adopted by the ICRP (1987) for indoor and outdoor exposure to ²²²Rn and the large variation in the other reported values, it does not seem very useful to retain separate values for the dose conversion coefficients indoors and outdoors.

It is not possible to choose between the three major dosimetric models mentioned on the basis of scientific arguments (BEIR IV, 1988); so, with the present state of scientific knowledge, the differences in the dosimetric results reflect the uncertainty inherent in such model calculations.

As reference values for the various dose conversion coefficients, it is probably best to take averages of the values in Tables 6.3 and 6.5, with as an uncertainty interval the mathematical dispersion in these values. This uncertainty interval indicates the domain within which the dose conversion coefficient for typical situations will lie with a probability of about 70%. For individual situations, however, the uncertainty interval will be greater. A summary of the dose conversion coefficients is given in Table 6.5.

Table 6.5. Conversion coefficients for the dose equivalent, in mSv a⁻¹ per Bq m⁻³ EEC exposure (in parentheses, effective dose equivalent in mSv WLM⁻¹). The stated figures are rounded off values

	²²² Rn	²²⁰ Rn
Dose equivalent in bronchial epithelium	2±1 (140±70)	7±3 (34±14)
Dose equivalent in pulmonary tissue	0.24+0.06 (17±5)	2±1 (11±6)
Effective dose equivalent to whole body	$0.14\pm0.05(10\pm4)$	$0.6\pm0.2(3\pm1)$
Dose ratio K (house/mine)	1±0.2	

As a reference value for the conversion coefficient of 222 Rn for the effective dose equivalent in typical situations, a value of 0.14 ± 0.05 mSv a^{-1} per Bq m⁻³ EEC, or 10 ± 4 mSv WLM⁻¹, can be used. For 220 Rn, this reference value is 0.6 ± 0.2 mSv a^{-1} per Bq m⁻³ EEC, or 3 ± 1 mSv WLM⁻¹. In individual cases, larger variations must be considered. These dose conversion coefficients refer to adults. For children under 10 years, values 1.5 times higher can be taken, if desired.

A dose conversion coefficient of 0.06 mSv a⁻¹ per Bq m⁻³ can be used in the estimation of the effective dose equivalent on the basis of the exposure to ²²²Rn gas (not EEC). The uncertainty in this value is not known but will probably be of the same order as for the other conversion coefficients, that is, plus or minus 30%, because this value too is based on similar model calculations.

The dose ratio K of 1 ± 0.2 implies that, for a given ²²²Rn exposure, the dose received by a person in a home is very nearly the same as the dose to a miner. Therefore, the epidemiological data from miners may, as far as this matter is concerned, also be applied to the general population.

6.1.5 Animal studies

Studies of the effects of inhaled ²²²Rn and daughters on experimental animals have been conducted mainly in the United States (dogs, rats; University of Rochester, Pacific Northwest Laboratory - PNL) and France (rats; Compagnie Générale des Matières Nucléaires - COGEMA). In the Netherlands, investigators at the ITRI-TNO exposed rats to ²²²Rn and progeny. In rats exposed to 800 WLM or less, no lung tumours were noted over an observation period of about 1.5 years.

Lung tumours have been induced in rats at relatively low exposures (20 WLM) (Chameaud et al., 1984). Dogs have not been exposed to such low WLM values. Lung tumours have been observed in dogs for exposures of 600 WLM and above (Cross et al., 1982). Risks coefficients for life-span shortening due to induced lung tumours are summarized in Figure 6.4. Lung-cancer incidence in laboratory animals increases with decreasing rate of exposure for fixed cumulative exposure. This finding has yet to be confirmed in studies of exposed underground miners.

Experiments in rats found that the activity concentrations in lung tissue and adipose tissue were similar after inhalation of ²²²Rn and progeny. Other organs had a lower activity, about a factor of 10 (see also 6.1.3). Since the mammary gland contains adipose tissue, the risk of mammary-tumour induction cannot be ruled out. In experiments at COGEMA, France, an increased frequency of mammary tumours was observed in female rats after exposure to ²²²Rn (pers. commun.).

Factors influencing the carcinogenic effectiveness of ²²²Rn and progeny exposure in laboratory animals are summarized in Table 6.6.

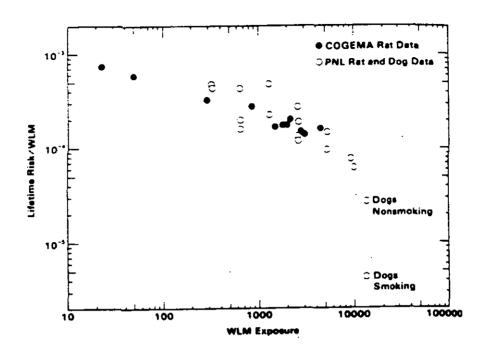


Figure 6.4. Lifetime risk coefficients for ²²²Rn-daughter exposure for PNL and COGEMA rat and dog data (BEIR IV, 1988).

Factor	Effect on respiratory tract tumour incidence
Cumulative exposure	Increases linearly with exposure
Exposure rate	Increases with decrease in exposure rate (about 200 to 400% increase from 500 to 50 WLM per week)
Unattached fraction, f	
Equilibrium factor, F	Increases with increase in disequilibrium (about 30% increase per WLM exposure from 0.4 to 0.1 equilibrium)
Concomitant exposure to cigarette smoke	 Decreases if smoking alternates with radon exposures on same day Increases if smoking follows cumulative radon exposures
	 No effect if smoking precedes cumulative radon exposures

Results of a few studies on lung cancer induction from inhalation of alpha-emitters attached to aerosols have also been reported in the literature. C57BL/6J mice were exposed by inhalation to aerosols of ²³⁹PuO₂ every other month (Lundgren et al., 1987). A total of six exposures were given over a period of 10 months. Other groups of mice received a single inhalation exposure. Groups of mice with similar cumulative alpha doses to the lung had 3.4 to 4.4 times greater incidence of lung tumours (adenomas and adenocarcinomas) when the dose was protracted by the repeated inhalation exposures as compared with mice that received a single exposure.

Published results from experiments investigating the effect of chronic exposure to alpha radiation are contradictory. Protraction of the alpha dose to the lungs of rats repeatedly exposed by inhalation at either weekly intervals for 9 to 22 weeks or monthly intervals for 3 months did not increase the carcinogenic effects of ²³⁹Pu but may have altered the type of induced tumours (Sanders and Mahaffey, 1981). In Syrian hamsters, protraction of exposure over 120 days was slightly more carcinogenic at lower total lung doses (0.24 Gy), but slightly less carcinogenic at higher doses (2.4 Gy), than exposure limited to a 10-day interval (Little et al., 1985).

Although some results are contradictory, it is clear that protraction of relatively low doses of alpha radiation does not reduce the risk of tumour induction (see Figure 6.4).

6.1.6 Epidemiological studies

Epidemiological studies have shown that chronic exposure to relatively high radon levels induces lung cancer. The following information is of relevance:

- 1. A direct relationship between chronic exposure to relatively high ²²²Rn levels and lung cancer occurrence can be derived from data from miners working underground in uranium and other mines;
- 2. The relationship between radon exposure and lung cancer occurrence can be derived indirectly from risk data for low LET radiation from studies of the survivors of atomic bombings in Hiroshima and Nagasaki and patients treated with radiation. On the basis of the risk coefficients derived from these data, the absorbed dose determined by using a dosimetric model, and the quality factor for alpha radiation from radon and its radioactive progeny, the risk from radon exposure can be estimated:
- 3. Because of the large differences in background ²²²Rn concentrations, it is possible, in principle, to derive the lung-cancer risk from a correlation study on population groups exposed to different natural ²²²Rn levels. This would be the most direct approach. In practice, however, these studies do not yield a statistically significant correlation.

The three different categories of populations exposed to radiation are discussed separately below.

Underground miners

Radiogenic lung cancer is the oldest type of radiation-induced malignancy known. It was recorded as early as the 15th and 16th centuries among miners in the Schneeberg and Jachymov (formerly Joachimsthal) areas in the "Ore Mountains" (Erzgebirge). This so-called "Schneeberger Krankheit" was diagnosed as lung cancer in 1879. Its possible association with radon was suggested about 60 years ago, when the high radon levels in mines of this region were discovered. However, the real cause of this disease, the inhalation of radon daughters, was not recognized until the 1950s, when the first results of dosimetric calculations became available.

Since then, numerous studies on ²²²Rn-exposed underground miners have shown that the

observed lung cancer frequency among these miners was significantly higher than in control populations. The results of these studies have been summarized by the ICRP (1987) and the BEIR IV Committee (1988). For estimating the risk associated with radon exposure, the cited reports contain data on uranium miners in Colorado (USA), Bohemia (Czechoslovakia), Ontario (Canada), Eldorado Beaverlodge (Canada), France and the ironore miners in Malmberget (Sweden). The characteristics of these data on miners are summarized in Tables 6.7 and 6.8 (BEIR IV, 1988; ICRP, 1987). No data on individual exposures are available from the French study. The ICRP Task Group used the published reports in its risk analysis, in contrast with the BEIR IV Committee, who used the raw data and did its own model calculations, further worked out in Chapter 7.

Table 6.7. Characteristics of the four miner groups analyzed by the BEIR IV Committee (1988)

Cohort	Study period	Lung cancer deaths (n)	Person-years at risk
Eldorado	1950 - 1980	65	114,170
Ontario	1955 - 1981	87	218,810
Malmberget	1951 - 1976	51	27,397
Colorado (all exposed)	1951 - 1982	256	73,642
Colorado (< 2000 WLM)	1951 - 1982	157	66,237

<u>Table 6.8.</u> Basic data for epidemiological studies on uranium miners (from ICRP, 1987)

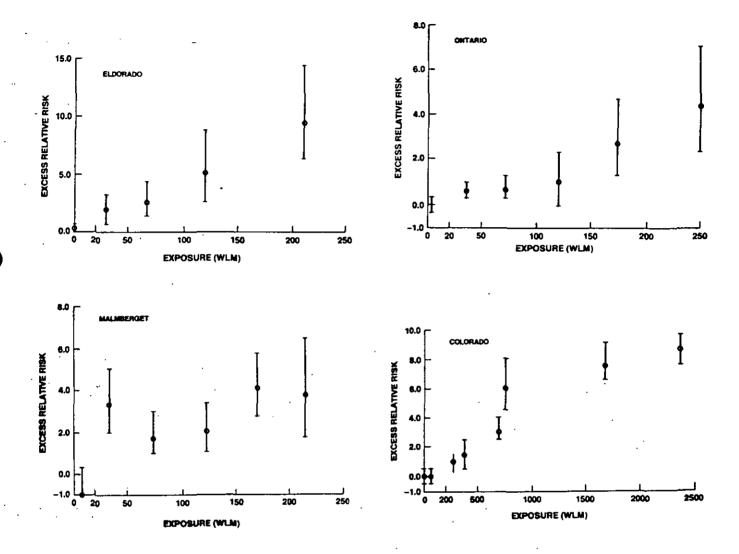
Quantity	Colorado a	Bohemia b) CSSR	Ontario c) Canada	France d)
	1950-1977	1948-1975	1955-1981	1947-1983
Initial number				
of miners	3,366	2,433	~ 13,400	1,957
Average follow-up	,			
period per miner (years)	19	26	15	25.9
Surviving fraction at end				
of follow-up (%)	72		~ 80	81
Median age at start of				
uranium mining (years)	30	35-40	~ 25	~ 30
Average working period in				
uranium mines (years)	9	10	~ 2	11.4
Number of person-years				
at risk (PYR)	62,556	~ 60,000	202,795	50,784
Mean cumulated exposure	•			
(WLM)	820	310	60±25	
Fraction of chronic				
cigarette smokers (%)	~ 70	~ 70	50-60	~ 70
Number of lung cancer cases	, •	, -		
during follow-up: observed	194	~ 250	82	36
expected		~ 50	57	18.8
excess	154	~ 200	25	17.2
Relative risk				- · • -
(observed/expected cases)	4.8	~ 5.0	1.45	1.9

a) White miners only

b) Study group A only; the total group involved 4,363 miners

c) Only uranium miners without prior gold mining experience

d) Exposure data not available



<u>Figure 6.5.</u> Excess relative risk and 67% confidence limits for exposure of: a: Eldorado uranium miners at Beaverlodge, Saskatchewan, Canada

- b: Uranium miners in Ontario, Canada
- c: Iron-ore miners in Malmberget, Sweden
- d: Uranium miners on the Colorado Plateau, USA

Exposure-response relationship

The ICRP (1987) analyzed the dose-response function for several epidemiological studies (Figure 6.5). For the Colorado miner data, a linear response yielded the best mathematical fit below a cumulative exposure of about 1000 WLM. Also, in the studies of the uranium miners in Czechoslovakia and in Ontario, the presumption of linearity, without threshold, cannot be rejected at the 5% significance level. The other studies are inadequate for the purpose of deriving a dose-effect relationship. The lowest cumulative exposure at which a statistically-significant excess risk has occurred is 50-100 WLM for the Czech and Ontario miners.

Atomic bomb survivors and patients receiving radiotherapy

Exposure to external radiation can induce lung cancer. This can be inferred from data on the atomic bomb survivors in Hiroshima and Nagasaki and patients with ankylosing spondylitis (Bekhteriv's disease, arthritis of the spine) in the United Kingdom. The most important data are listed in Table 6.9. An important difference between the Japanese and British cohorts is that 25 years after exposure to radiation, the lung cancer risk had returned to the background value in the British cohort, whereas despite a slight decrease, there was still an excess risk in the Japanese cohort. The populations of Hiroshima and Nagasaki were acutely exposed to mainly gamma radiation and, to a lesser extent, neutrons. In contrast, the British patients were irradiated non-uniformly and received a fractionated X-ray dose. A recent summary is given by the UNSCEAR (1988) and BEIR V Committee (1990). The risk estimates based on mathematical models are presented in Chapter 7.

Table 6.9. Data from cohorts exposed to external radiation

Atomic bomb survivors Hiroshima and Nagasaki DS86 cohort	Ankylosing spondylitis patients
1950-1985	1935-1982
75,991	14,106
638	224
5,936	727
1945	1935-1954
2,185,000	184,000
	184.5
	1.79 Gy
individual	collective < 0.01
59	17
acute whole body	<pre>fractionated non-uniform, part of body</pre>
	-
28.8	13.0
0 - > 90	> 15
0.63	0.12
10	5
	Hiroshima and Nagasaki DS86 cohort 1950-1985 75,991 638 5,936 1945 2,185,000 individual 59 acute whole body 28.8 0 -> 90 0.63 10

Populations exposed to enhanced natural ²²²Rn levels

Although the radon exposure in different regions of the world shows large differences, it has not (yet) been possible to derive directly from this what the health effects are.

Epidemiological studies in the United States (Hickey et al., 1981), in which air pollutants were also considered, found that natural radiation (of cosmic and terrestrial origin) was negatively correlated with lung cancer incidence.

An epidemiological case-control study (Stockwell et al., 1988) was conducted in 53 counties in Florida, USA: 18 counties with potentially elevated levels of indoor radon and 35 without elevated indoor radon levels served as the comparison group. A 25% increase in lung cancer risk was observed among residents of the three counties with the highest potential for elevated levels of radon, and no increased risk was observed among the remaining 15 counties with elevated radon levels.

A survey in England and Wales (Haynes, 1988) found a negative correlation between mean radon concentration in homes and lung cancer mortality. In Cornwall and Devon, with the highest domestic radon concentrations, the number of lung cancer cases was comparable to that in the rest of Britain.

That these direct assessments at enhanced natural radiation levels do not yield statistically significant results is possibly due to the predominant role of smoking in the incidence of lung cancer. Furthermore, an indoor exposure of 0.2 WLM per year, an average value in most countries, corresponds to a cumulative lifetime exposure of 10 to 20 WLM, which is a factor of 2 to 5 lower than the lowest level at which a statistically significant increase in lung cancer frequency has been observed among uranium miners.

Henshaw et al. (1990) demonstrated a significant correlation between radon exposure in the home and leukaemia incidence in children. A significant correlation with other childhood cancers is also suggested. Animal experiments have shown that ²²²Rn accumulates in adipose tissue and from there may expose surrounding organs and tissues (see 6.1.3).

However, the epidemiological studies among the general public published to date do not enable conclusions to be drawn about the effects associated with relatively low ²²²Rn exposures. Recently initiated case-control studies might provide more information in the coming years.

6.1.7 Interaction between radon and smoking

Exposure to radon and its daughters and cigarette consumption are each associated with lung cancer. The BEIR IV Committee (1988) analyses show that lung cancer risk associated with exposure to radon progeny depends on cumulative exposure, age at first exposure, and time elapsing after cessation of exposure. The actual biological relationship is undoubtedly more complex than described in the statistical model and may be influenced by other factors that cannot be fully evaluated with the available data. Moreover, the association of tobacco consumption with lung cancer is also complex and depends on the number of years of smoking, the number of cigarettes smoked per day, type of tobacco product, method of inhalation, and for former smokers the number of years since smoking was stopped. In assessing the combined effects of cigarette smoking and radon progeny we should account for the individual patterns of effects from both insults. Other aspects of the combined exposure may also be important, for example, the effect of sequencing of exposures and the degree of their overlap in time.

However, the studies of combined exposures have usually considered only cumulative WLM and duration or intensity of cigarette use. Nevertheless, risk models can be useful in establishing trends. Two commonly used risk models are the additive and the multiplicative model.

The additive model assumes that the number of excess lung cancer cases resulting from the combined exposure is the sum of the excess number of lung cancer cases due to the separate, individual effect of smoking and radon exposure. This model therefore assumes that there is no synergism between the two exposures. In the multiplicative model, it is assumed that the relative risk from the combined exposure is the product of the relative risks from the separate exposures. The relative risk is the ratio of the number of lung cancer cases observed in an exposed population to the number of cases expected in a non-exposed, but otherwise comparable, population.

A clear pattern of risk among studies of miners exposure to radon and tobacco smoke has not yet emerged. Although the BEIR IV analysis (1988) of the Colorado Plateau uranium miners favour the multiplicative model, they also support sub-multiplicative and supra-

multiplicative relationships. The largest study of the issue by Whittemore and McMillan (1983) also indicates a multiplicative interaction. The BEIR IV Committee's analysis of the Japanese atomic bomb survivor data show that for these data, neither the additive nor the multiplicative model can be rejected on statistical grounds. The relevance of these studies of atomic bomb survivors to the interaction of radon and smoking in their relationship to lung cancer induction, however, must still be determined.

The BEIR IV Committee (1988) concluded that the interaction between radon and smoking is strongly synergistic. The BEIR V Committee (1990) considered the same data inadequate for analysis of interaction effects. Also, the results of the few available animal studies (see Table 6.6) do not lead to a firm conclusion on the form of the interaction between radon and smoking. Extensive studies in laboratory animals might resolve this issue.

6.2 ECOTOXICITY

6.2.1 Aquatic and terrestrial organisms

Apart from the animal studies already discussed (see 6.1.5), there are no experimental data available on the detrimental effects of radon on aquatic and/or terrestrial organisms. However, a rough dose estimate can be made for both.

With regard to aquatic organisms, the radon concentration in the organism will, to a first approximation, be the same as the concentration in the surrounding water. This need not be true for the daughter products, because they can be selectively taken up or excreted. In the absence of further data, however, the same concentration inside and outside the organism will also be assumed for the daughters.

The 222 Rn concentration in the North Sea is about 1.5 Bq m⁻³ (Blaauboer et al., 1989). Assuming that 222 Rn is in full radioactive equilibrium with its daughters (including 210 Po, so that on decay of one atom of radon to stable 206 Pb, 3.9×10^{-12} J of alpha energy is ultimately released), the emission is found to be 1.5 (Bq m⁻³) x 3.9×10^{-12} (J) x 10^{-3} (m³ kg⁻¹) x 32×10^{6} (s a⁻¹) = 0.19 μ Gy a⁻¹ of alpha radiation due to 222 Rn. Because the radon concentration in rivers, lakes and ponds need not be the same as the concentration in the North Sea, the dose rate to organisms in these water bodies can differ.

An analogous estimate can be made for terrestrial organisms such as worms. Assuming full radioactive equilibrium in soil (water), including ²¹⁰Po, and a ²²²Rn concentration of 1x10⁵ Bq m⁻³ (Blaauboer et al., 1989), the alpha dose in water is calculated to be 12 mGy a⁻¹.

Animals living in burrows, such as moles, are also exposed to radon (daughters) through inhalation, as well as by ingestion. Pertinent data are not available, but it is not inconceivable that the lung dose to these animals is considerable. Assuming a concentration in tunnels and holes 100 times lower than in the soil itself (about 50,000 Bq m⁻³ soil gas; see 5.1), and using a dose conversion coefficient for lung tissue of the same order as for humans (i.e. about 1 mSv a⁻¹ per Bq m⁻³), this yields a dose of 0.5 Sv a⁻¹.

6.2.2 Bioaccumulation

Because of the short half-lives of ²²²Rn and ²²⁰Rn, bioaccumulation will not occur. Only the long-lived daughters such as ²¹⁰Pb might accumulate, but few pertinent data are available. A case of accumulation of radon daughters, where humans might receive considerable doses, is the accumulation in the tobacco plant of ²¹⁰Pb deposited from the

atmosphere, from which ²¹⁰Po is formed by decay (see 3.1). These nuclides can enter the lungs with inhaled tobacco smoke and so cause exposure of the bronchial epithelium. Cohen et al. (1980) measured the ²¹⁰Po activity concentration in small pieces of bronchial epithelium from deceased smokers, ex-smokers and a non-smokers (seven in all), and found that the activity in older smokers is, as expected, higher than in non-smokers. On the basis of this study, the NCRP Report 93 (1987) calculated an upper limit for the annual effective dose equivalent to smokers of 13 mSv.

According to the NCRP, the risk associated with the upper limit of 13 mSv a^{-1} would, in a lifetime, amount to about 1%, or only a portion of the total risk of lung cancer to smokers (3-9%). The estimate of this upper limit is based on one specimen of lung tissue of an elderly smoker, which showed a much higher activity than the other samples examined. On the basis of the mean measured activity of the samples, however, a dose can be calculated which is lower than the upper limit of 13 mSv a^{-1} adopted by the NCRP by at least a factor of 500, that is, a few tens of μ Sv per year. Such a dose is much smaller than the natural radiation dose and, moreover, yields an excess risk which is negligible compared with the "normal" risk from smoking.

6.3 TOXICITY TO LIVESTOCK

Livestock kept in stables are exposed to radon (daughters) by inhalation. Data on radon concentrations in stables are not yet available, but in view of the fact that such spaces are often ventilated, high radon levels are not expected. Taking a dose conversion coefficient for livestock of the same order as for humans, it then also does not seem likely that the doses to stabled livestock will be higher than the average dose to man, that is, approximately a few mSv a⁻¹. Cattle at pasture can also be exposed to radon by ingestion, as well as by inhalation, but no data are available. Because of the low outdoor radon concentrations, the dose from inhalation will be negligible.

6.4 SUMMARY AND CONCLUSIONS

Epidemiological studies of underground miners and animal experiments have shown that exposure to the element radon induces lung cancer. There are also indications that radon can cause other forms of cancer. The principal mode of entry of ²²²Rn, ²²⁰Rn and their daughters into the body is inhalation. After inhalation, a proportion is taken up into the blood and distributed in the body. The effective dose equivalent can be estimated by using a mathematical model. Dose conversion coefficients represent the ratio between the exposure to ²²²Rn, ²²⁰Rn and their daughters in air, and the respective dose equivalent in the lung. For children the lung dose per unit exposure might be up to a factor of 2 higher than for adults.

The main conclusions of this chapter are:

- 1. For calculating the dose equivalent, different values for the alpha radiation quality factor have been recommended. International agreement on the value to be assigned to the quality factor is needed.
- 2. There are indications (epidemiology, animal experiments) that ²²²Rn can cause cancers other than lung cancer. Research into the distribution of radioactivity in organs and tissues, particularly adipose tissue, and into the risk, especially of leukaemia and mammary-tumour induction, following ²²²Rn exposure is recommended.

- 3. Evaluation of various lung models on the basis of model parameters does not result in a preference for a particular model. At present, the use of different models gives an indication of the uncertainty in the results.
- 4. For the present, the value of K, the proportionality factor for conversion of the risk coefficient obtained for underground miners to the risk coefficient for members of the general population may be equated to 1. The uncertainty in this value is about 20%.
- 5. For the dose conversion coefficient for typical situations, i.e. the effective dose equivalent (rate) per unit continuous exposure, a value of 0.14 ± 0.05 mSv a⁻¹ per Bq m⁻³ EEC, or 10 ± 4 mSv WLM⁻¹, can be used for ²²²Rn. For ²²⁰Rn, this conversion coefficient is 0.6 ± 0.2 mSv a⁻¹ per Bq m⁻³ EEC, or 3 ± 1 mSv WLM⁻¹. For individual exposures, larger variations must be considered.
- 6. The epidemiological studies among the general population published to date do not allow conclusions to be drawn about the health effects associated with relatively low ²²²Rn exposures.
- 7. For the relationship between radon exposure and lung cancer induction, a linear dose-response pattern without threshold is assumed.
- 8. A firm conclusion on the form of the interaction between radon exposure and smoking cannot be drawn on the basis of data from studies on miners. The analyses by the BEIR IV Committee suggest a strong synergism, but BEIR V considers the data too weak to support this. The few animal data currently available also do not allow firm conclusions to be drawn. Extensive studies on laboratory animals might provide an answer.
- 9. No data are available on possible detrimental effects of radon and radon daughters on aquatic and terrestrial organisms, and livestock.

7 RISKS

Exposure of humans to radon and its daughters, primarily originating from the soil and building materials, is mainly by inhalation. In addition, external exposure of the entire body is caused by gamma radiation arising from short-lived radon daughters contained in soil and building materials.

In this chapter, estimates of the risk resulting from exposure to radon are listed and evaluated. The risk calculation for ²²²Rn is based on data from epidemiological studies on miners, that for ²²⁰Rn on lung dosimetry. A check on the risk estimate for ²²²Rn is made by extrapolation of data from survivors of atomic bombings in Hiroshima and Nagasaki. The influence of exposure conditions on risk coefficients (indoors, outdoors) is examined using dosimetric models, followed by attention to sensitive and critical groups. A risk estimate for the Dutch population is given. Possible psychological effects of ionizing radiation are also considered. Finally, the effects on aquatic and terrestrial organisms, and livestock, are briefly outlined.

7.1 HUMAN RISKS

Risk coefficients for ²²²Rn exposures in houses can be derived from studies on miners (BEIR IV, 1988; ICRP Publication 50, 1987). Risk coefficients can also be derived from studies on populations exposed to other types of radiation, where it is necessary to use a dosimetric model and the quality factor for alpha radiation for conversion to radon exposure. In general, lifetime lung-cancer mortality is used as the risk criterion, and the relative risk model may be applied to calculate the number of lung cancer deaths due to a given exposure.

7.1.1 Risk evaluations by BEIR, ICRP and UNSCEAR committees

BEIR IV analysis of miner data

The BEIR IV Committee (1988) statistically analyzed the raw data from four cohorts of underground miners using a modified relative risk model:

$$\mathbf{r}(t) = \mathbf{r}_0(t)[1 + 0.025 \ \gamma(t)(\mathbf{W}_1 + 0.5 \ \mathbf{W}_2)] \tag{7.1}$$

where r(t) is the lung-cancer mortality rate at age t; $r_0(t)$ is the background lung-cancer mortality rate at age t; W_1 is the cumulative radon exposure in WLM from 5-15 years before age t; and W_2 is the cumulative exposure 15 years or more before age t. The division of the exposure into W_1 and W_2 accounts for a decline in the relative risk with time since exposure. The function $\gamma(t)$ accounts for the increased risk for ages less than 55 years. The assumed latency period is five years.

The BEIR IV (1988) Committee's risk projections are based on the assumption that the relative risk is the same for males and females. Furthermore, the Committee has assumed that the lung cancer risk per unit of exposure is the same in a mine as in a home. BEIR IV has estimated the lifetime risk of lung-cancer mortality due to a lifetime exposure to ²²²Rn and its decay products to be 350x10⁻⁶ WLM⁻¹.

The ICRP analysis of miner and atomic bomb survivor data

In Publication 50 (1987) the ICRP analyzed the miner data using both a constant absoluterisk model and a constant relative-risk model. The relative-risk model assumes a latency

period of 10 years. The latency period is also 10 years in the absolute-risk model, but the risk does not start until age 40. In the case of a single exposure, the constant absolute-risk model assumes that, after the latent period, the annual excess lung-cancer mortality risk remains constant. The constant relative-risk model assumes that, during the time at risk, the ratio of the excess risk to the background age-specific risk of lung-cancer mortality is constant.

In the BEIR IV (1988) notation, the absolute risk model is represented by:

$$r(t) = r_0(t) + a_1 W_3 (7.2)$$

and the constant relative-risk model by:

$$r(t) = r_0(t) (1 + r W_3)$$
 (7.3)

where W₃ is the cumulative exposure after the latency period.

The constants a_1 and r can be derived from data on miners. The ICRP found $a_{1,miners} = 10x10^{-6} a^{-1} WLM^{-1}$; $r_{miners} = 0.010 WLM^{-1}$.

In addition to being exposed to radon, the underground miners were also exposed to long-lived radionuclides, gamma radiation, and other carcinogenic or synergistic non-radioactive dusts and vapours present in mine air. The ICRP has estimated that about 20% of the overall excess risk might be attributed to sources other than ²²²Rn daughters, corresponding to a multiplicative correction factor of 0.8.

The lung cancer data for underground miners refer only to males. Therefore, to evaluate the risk of radiation-induced lung cancer to females, other data have to be utilized, such as the Japanese atomic-bomb survivor data. On the basis of the observations on this group of survivors, the ICRP has estimated the risk of radiation-induced lung cancer to be the same for males and females; the same age-specific relative risk coefficients are assumed for both sexes. On the basis of the atomic-bomb survivor data, the ICRP has assumed a mean relative risk coefficient for the 0-20 age group which is three times larger than for adults. These corrections give a population risk coefficient of:

$$r_{population} = 0.019 \text{ WLM}^{-1} \text{ for } t < 20 \text{ years, and } 0.0064 \text{ WLM}^{-1} \text{ for } t \ge 20 \text{ years.}$$

For the overall uncertainty of the risk coefficients, the ICRP (1987) gives a range from 0.3 up to twice the values stated.

The ICRP Commission believes that the relative-risk model enables a more reliable risk projection than an absolute-risk model. The ICRP (1987) has calculated a risk of 230x10⁻⁶ WLM⁻¹ for a reference population which is representative of the whole world population and chronically exposed to ²²²Rn.

The BEIR V Committee (1990) applied the ICRP calculation to the 1980 population of the United States and obtained a risk of 360x10⁻⁶ WLM⁻¹.

If the above corrections are applied to the absolute-risk model, the risk coefficient for the population over 40 is:

$$a_{1,population} = 7x10^{-6} a^{-1} WLM^{-1}$$
 (7.4)

For a population with a life expectancy of 73 years without 222Rn exposure, chronic

exposure to ²²²Rn yields an excess lifetime risk of lung-cancer mortality of 150x10⁻⁶ WLM⁻¹.

UNSCEAR 88 analysis of lung cancers in Japanese atomic bomb survivors

UNSCEAR (1988) has also made an extensive analysis of data from the Japanese atomic bomb survivors. Both absolute-risk and constant relative-risk models have been applied. The excess risk of a fatal lung cancer is 1.68×10^{-4} a⁻¹ Gy⁻¹ based on the absolute-risk model and 0.63 Gy⁻¹ on the relative-risk model. For the Japanese population, this yields a lifetime risk of death from a radiation-induced lung cancer of 59×10^{-4} Gy⁻¹ (90% confidence interval: $(34-88)\times 10^{-4}$ Gy⁻¹) on the basis of the absolute-risk model and of 151×10^{-4} Gy⁻¹ (90% confidence interval: $(84-230)\times 10^{-4}$ Gy⁻¹) under the relative-risk model. These values are for exposure to 1 Gy low-LET radiation, with a high dose rate.

In the report "Gezondheidsschade Mens" ("Human Health Damage") (Kal et al., 1988), following the approach of the UNSCEAR 1988 Report and the ICRP Publication 50 (1987), the absolute and relative risk coefficients derived for the population of Japan have been transferred to the Dutch population. The lifetime risk of a fatal radiation-induced lung cancer is then 54x10⁴ Gy⁻¹ for the absolute-risk model and 388x10⁴ Gy⁻¹ for the relative-risk model. The large difference in risk estimates resulting from the relative-risk model is caused by the large differences in baseline lung-cancer mortality rates between the Japanese and the Dutch populations. The BEIR V Committee (1990) states that the relative risk model possibly describes effects of age at exposure but that for transfer between populations additive risk coefficients must nevertheless be used. In the study by Darby et al. (1987) concerning British patients treated with X rays for ankylosing spondylitis, about 39.5 excess lung cancer deaths were observed five years after exposure to radiation in a total of 183,749 person-years at risk and, according to Lewis et al. (1988), a mean lung dose of 1.79 Gy (standard deviation of 1.57 Gy). This yields an absolute-risk coefficient of 1.20x10⁴ a⁻¹ Gy⁻¹ and an excess relative-risk coefficient of 0.12 Gy-1. As previously mentioned, the risk coefficients derived from the Japanese atomic bomb survivors are 1.68x10⁻⁴ a⁻¹ Gy⁻¹ and 0.63 Gy⁻¹. This indicates that, with regard to lung cancer, it is probably better to transfer the additive risk coefficients between the Japanese and the British population than the relative-risk coefficients.

A preliminary analysis of the miner data by UNSCEAR (1988) suggests no large changes in relation to the lifetime lung-cancer mortality risk of (150-450)x10⁻⁶ WLM⁻¹ estimated by UNSCEAR in 1977.

BEIR V analysis of lung cancers among atomic bomb survivors and ankylosing spondylitis patients

The BEIR V Committee (1990) assessed the risk using a modified relative risk model. For a given radiation dose d, the individual age-specific fatal cancer risk $\gamma(d)$ is:

$$\gamma(d) = \gamma_0 \left[1 + f(d) - g(\beta) \right] \tag{7.5}$$

where γ_0 denotes the age-specific background risk due to a specific fatal cancer for an individual of a given age, sex and birth cohort; and f(d) represents a linear or linear-quadratic function of the dose d. This equation enabled the BEIR V Committee to implicitly indicate the dose reduction factor. In general, the excess risk function, $g(\beta)$ will depend upon a number of parameters, for example, sex, attained age, age at exposure and time since exposure. It may be noted that this model is equivalent to an additive risk model, because the function $g(\beta)$ is dependent on age at exposure and sex.

The BEIR V Committee's preferred model for lung cancer mortality, after a 10-year latency, is as follows:

$$f(d) = \alpha_1 d \tag{7.6}$$

$$g(\beta) = \exp [\beta_1 \ln (T/20) + \beta_2 I(S)]$$
 (7.7)

where T is time after exposure and I(S) is 1 if female and 0 if male. The estimated parameter values with their standard errors in parentheses are:

$$\alpha_1 = 0.636 (0.291)$$

 $\beta_1 = -1.437 (0.910)$
 $\beta_2 = 0.711 (0.610)$

The coefficient for time after exposure, -1.437, means that the relative risk will decrease by a factor of about 5 over the period of 10 to 30 years after exposure (a decline in the relative risk is also observed in the ankylosing spondylitis patients). The "sex" term means that the relative risk is twice as high for females as for males. However, owing to females much lower baseline rates, the observed excess risks are similar. When a term for "age at exposure" was added to the model, its estimated value was sufficiently close to zero as to have no influence on the risk estimate.

Using the data on life expectancy of the USA population and the cancer mortality statistics of 1980, the BEIR V Committee (1990) found an average lifetime excess risk of lung cancer mortality of 190×10^4 Sv⁻¹ for males and 150×10^4 Sv⁻¹ for females.

Estimates of the mortality risk due to exposure to ²²²Rn and daughters, based on UNSCEAR 88 and BEIR V data

Under the relative-risk model, the lifetime mortality risk estimated from the Japanese atomic-bomb survivor data by UNSCEAR (1988) is $150x10^{-4}$ Sv⁻¹, and that by the BEIR V Committee (1990) is $190x10^{-4}$ Sv⁻¹ for males and $150x10^{-4}$ Sv⁻¹ for females. These values refer to exposure to low-LET radiation at a high dose rate. In order to determine the risk at low dose rates, a dose-rate reduction factor of about 2 must be introduced (Kal and Jansen, 1990), resulting in a mortality risk of about $80x10^{-4}$ Sv⁻¹. The dose equivalent to the bronchial epithelium can be estimated at about 140 mSv WLM⁻¹, the dose equivalent to the pulmonary tissue at 17 mSv WLM⁻¹ (Table 6.3), and thus the dose equivalent to the whole lung at about 80 mSv WLM⁻¹. The excess lifetime risk of lung-cancer mortality then follows from the mortality risk derived from the Japanese atomic bomb survivors (80x10⁻⁴ Sv⁻¹) and the lung dose equivalent (80 mSv WLM⁻¹) at approximately 640x10⁻⁶ WLM⁻¹.

Radon-220 (thoron)

The risk from inhalation of ²²⁰Rn and its daughters cannot be derived from epidemiological studies among miners: the dominant risk in mines is ²²²Rn and its daughters. It is possible, however, to make an estimate of the risk by using lung models. It was found that with respect to the risk per unit of activity, ²²²Rn and ²²⁰Rn are not comparable. ICRP (1987) argues that the doses to tissues outside the respiratory tract from inhaled ²²²Rn and progeny are negligible compared with the dose to the lung. For inhaled ²²⁰Rn and progeny, however, the transfer of ²¹²Pb also leads also to significant doses to tissues outside the lung, particularly bone surfaces, bone marrow, kidneys and liver. An effective dose

equivalent of 3 mSv per WLM is given in Table 6.5. According to ICRP Publication 26 (1977), the mortality risk from radiation-induced cancers is about 1.25% per Sv. In ICRP 60 (1991), the ICRP Commission proposed to increase the mortality risk to 5% per Sv for the whole population and to 4% per Sv for the working population (it can be mentioned that the tissue weighting factors used for calculating the effective dose equivalent have been modified). This yields a mortality risk of 150×10^6 WLM⁻¹ (28×10^{-6} a⁻¹ Bq⁻¹ m³) from ²²⁰Rn and its daughters to the general population. If the same method is followed for exposure to ²²²Rn and daughters, the mortality risk is found to be 500×10^{-6} WLM⁻¹ (7×10^{-6} a⁻¹ Bq⁻¹ m³).

External irradiation

Radon-222 and ²²⁰Rn in the ground and in building materials which are not released into the air can still give rise to external irradiation due to decay of the daughter products (see also 8.1.4). For the Netherlands, the annual gamma-ray effective dose equivalent received indoors has been calculated to be about 0.2 mSv (contributions from the ²²⁶Ra subseries, the ²³²Th series, and ⁴⁰K) (Ackers, 1985; 1989). It was assumed that the indoor occupancy factor is 0.80. If the ⁴⁰K contribution is not taken into account, the annual effective dose equivalent is 0.15 mSv. Not all the gamma radiation from the ²³²Th series arises from the short-lived decay products of ²²⁰Rn, but ²²⁸Ac also plays a role. No attempt was made to correct for this. With the indoor occupancy factor of 0.88 adopted in this document, the annual effective dose equivalent is 0.17 mSv.

UNSCEAR (1988) gives a kerma rate in air 1 m above the ground surface of 0.427 and 0.662 nGy h⁻¹ per Bq kg⁻¹ (wet weight) for the ²³⁸U and ²³²Th series, respectively. The average activity concentrations of ²²⁶Ra and ²³²Th are 25 and 35 Bq kg⁻¹ (see Table 5.1). In this report, it is assumed that all decay products of the ²³⁸U series are in radioactive equilibrium with their precursors, which means that the activity concentrations of ²²⁶Ra and ²³⁸U are the same. A portion of the exposure is due to ^{234m}Pa. Using a conversion factor of 0.7 Sv Gy⁻¹ from air kerma to effective dose equivalent, and an outdoor occupancy factor of 0.12, the annual effective dose equivalent for the Netherlands is estimated to be 20 μ Sv.

The mortality risk from radiation-induced cancers is taken to be 5% per Sv (0.05 Sv⁻¹) for the whole population (ICRP, 1991).

<u>Table 7.1.</u>	Comparison	of e	estima	tes	for	risk	of	fatal	lung-ca	ancer	due t	oa
	lifetime	expo	sure	to 2	²² Rn	and p	proge	eny, ba	ased on	miner	data	. •

Report	Year	Excess lifetime risk (10.6 a.1 Bq.1 m3)	of fatal lung-cancer (10.6 WLM.1)
BEIR IV	1988	4.9	350
ICRP	1987	2.1-3.2 (a) 5.0	150~230 (a) 360 (b)
NCRP	1984	1.8	130
BEIR III	1980	10	730
UNSCEAR	1977	2.1-6.3	150~450

⁽a) relative risk with ICRP reference population

Conclusions

Determination of the risk from exposure to ²²²Rn and progeny is more direct using the data obtained in epidemiological studies on miners than using mathematical models or data on the Japanese survivors of atomic bombings. The relative-risk model seems to give a

⁽b) relative risk with 1980 US population as in BEIR IV

better description of effects of age at exposure than the absolute-risk model. From the miner data, the ICRP (1987) and BEIR IV Committee (1988) derived on the basis of the relative-risk model a lifetime mortality risk from radiation-induced lung cancer of 230x10⁻⁶ WLM⁻¹ and 350x10⁻⁶ WLM⁻¹, respectively. Replacing the ICRP reference population by the USA population, as in BEIR IV, yields a lifetime mortality risk of 360x10⁶ WLM⁻¹. Considering the uncertainties involved, the agreement between the different methods is reasonable. These estimates are summarized in Table 7.1. The BEIR IV estimate replaces the 1980 estimate in BEIR III. The ICRP estimate, with the 1980 USA population as reference, seems to be more relevant to the Dutch situation than a world reference population. The lifetime risk of lung-cancer mortality due to inhaled ²²²Rn and progeny is thus estimated to be 350x10⁻⁶ WLM⁻¹ (4.9x10⁻⁶ Bq⁻¹ m³ a⁻¹), with an uncertainty of roughly a factor of 2. Combining this with the dose conversion coefficient for ²²²Rn for typical situations of 0.01 Sv WLM⁻¹ (see 6.4) results in a mortality risk due to ²²²Rn of 0.035 Sv-1. The risk of lung-cancer mortality due to ²²²Rn derived from the data on the Japanese survivors of atomic bombings is about 640x10⁻⁶ WLM⁻¹, and is not significantly different from the above-mentioned risk value of 350x10⁻⁶ WLM⁻¹, with an uncertainty factor of 2. The risk of mortality due to inhaled ²²⁰Rn and progeny cannot be derived from epidemiological data. Using the dosimetric approach, the mortality risk is estimated to be 150x10⁻⁶ WLM⁻¹ (28x10⁻⁶ Bq⁻¹ m³ a⁻¹), also with an uncertainty of roughly a factor of 2. Taking the dose conversion coefficient for ²²⁰Rn for typical situations of 0.003 Sv WLM⁻¹ (see 6.4), the mortality risk due to ²²⁰Rn is found to be 0.05 Sv⁻¹.

7.1.2 <u>Influence of exposure conditions</u>

The most reliable risk coefficients for lung cancer due to exposure to ²²²Rn and progeny are derived from epidemiological studies on miners. The exposure conditions for members of the general public in private homes, other buildings or elsewhere are different from those in a mine, due to differences in aerosol size distribution, attached versus unattached fraction, breathing rate and the presence of other carcinogens. Chapter 6 has explained why the risk coefficients per WLM for exposure in underground mines are applicable to exposure in other settings.

Correction factors according to ICRP

In addition to exposure to ²²²Rn and progeny, underground miners are also exposed to long-lived radionuclides, gamma radiation and other carcinogenic or synergistic non-radioactive dusts and vapours which are present in mines. The ICRP has estimated that about 20% of the total excess risk can be attributed to these sources, corresponding to a multiplicative correction factor of 0.8. The mean breathing rate of members of the public is lower than that of miners underground. This is particularly valid for the resting phase at night at home. This implies a lower dose per unit of exposure for members of the general population than for miners. This lower dose is partially compensated for, because in indoor air the fraction of unattached daughter atoms is somewhat higher and the activity median diameter (AMD) of the carrier aerosol of attached daughter atoms is probably somewhat smaller than in mine air. This would lead to an increase in the bronchial dose. The resulting correction factor for residence indoors is 0.8. According to the ICRP (1987), the bronchial dose incurred outdoors is virtually the same as that incurred in mines.

Correction factors according to BEIR IV

BEIR IV concludes that the risk per WLM to workers in underground mines is not

significantly different from the risk to adult members of the general population in houses and, therefore, no correction factors are recommended.

7.1.3 Sensitive and critical groups

In many countries, "spontaneous" lung-cancer risk is higher for males than for females. According to the BEIR IV Committee (1988), the relative risk is the same for both sexes and, therefore, the absolute excess of ²²²Rn-induced lung cancers can be higher in males than in females. In contrast, the ICRP (1987), UNSCEAR (1988) and BEIR V (1990) committees found that, per unit of exposure, the absolute excess of ²²²Rn-induced lung cancers was similar in males and females and that the relative risk differed. This is based on data from the Japanese atomic bomb survivors. After adjusting for the effects of smoking, the relative-excess risk as well as the absolute-excess risk did not differ significantly between the sexes.

The data on miners refer primarily to males and, therefore, cannot be used to derive a risk estimate for females because of a possible sex difference in susceptibility to radiation carcinogenesis. These data are also of no use for estimating the age-specific lung-cancer risk, because they cover only the age span of the working population. The ICRP Commission (1987) and BEIR IV Committee (1988) have assumed a larger dose per unit of exposure for young persons, based on dosimetric models. Furthermore, the ICRP Commission has estimated that, per unit exposure, individuals under 20 who are exposed have an approximately twice as high relative risk than individuals exposed at higher ages. This is based on the Life Span Study in Japan. Because the atomic bomb survivors who were exposed to ionizing radiation at an early age have now reached an age at which spontaneous lung cancer incidence begins to increase, the limited follow-up period lends a definite element of uncertainty to this figure. The UNSCEAR (1988) data, with a longer follow-up time, do not confirm this conclusion. For the age cohort of 0-10 years at the time of the bombings, the relative risk is even smaller than 1. The BEIR V Committee found little effect of age at exposure, but did observe a decline in the relative risk as time after exposure increased. According to the ICRP, the relative excess risk for young persons might be up to a factor of 4 higher than for adults.

The BEIR IV Committee found that the data on the combined effects of smoking and ²²²Rn exposure fit a sub-multiplicative risk model. They also estimated the risk of a ²²²Rn-induced lung cancer to be about 10 times greater for a smoker than for a non-smoker. In the risk evaluation in this document the cumulative exposure to radon throughout life (lifetime risk) is used, taking into account the effects of age and sex.

Subpopulations with relatively high exposure levels

The mean annual effective dose equivalent to members of the population from indoor and outdoor exposure to ²²²Rn is about 1 mSv in the Netherlands. Little is known about occupational exposures in working places with high ²²²Rn concentrations. With respect to exposure in the phosphate ore-processing industry, Lardinoye et al. (1982) have calculated the ²²²Rn concentration from the measured radium concentration in the various process flows of phosphoric acid production. The highest calculated concentration is 118 Bq m⁻³. Using the equilibrium factor F of 0.4, in indoor air, gives an equilibrium equivalent concentration of 47 Bq m⁻³. Working 400 h a⁻¹ at this concentration (Lardinoye et al., 1982) would result in an annual exposure of 3x10⁻² WLM or an annual effective dose equivalent of 0.3 mSv.

Crawl spaces of houses often have a high ²²²Rn concentration. Individuals who are occupationally exposed in these spaces can receive a relatively high dose equivalent.

Assuming an average ²²²Rn concentration of 180 Bq m⁻³ (Chapter 8), an equilibrium factor F of 0.4 (Chapter 5), and a 1000 h exposure per year in a crawl space (about 50% of the working time) would result in an annual effective dose equivalent of 1 mSv at an exposure rate of 0.1 WLM a⁻¹.

Critical groups have not yet been sufficiently identified, but identification is recommended. These groups may include persons occupationally exposed in crawl spaces, cellars or tunnels, and workers at hot springs.

Members of the population living in houses with a high ²²²Rn concentration also run a higher-than-average risk. As an example the risk calculation for living in a house with an equilibrium-equivalent radon concentration (EEC) of 100 Bq m⁻³ is given. Using an indoor occupancy factor of 0.70 this is EEC results in an annual exposure of 0.97 WLM or an annual effective dose equivalent of 9.7 mSv. However, dwellings with an annual mean concentration of 100 Bq m⁻³ (EEC) have not (yet) been found in the Netherlands (in the 894 living rooms studied, the highest EEC of ²²²Rn was 56 Bq m⁻³). The mean equilibrium equivalent of ²²²Rn concentration in Dutch single family dwellings is 12 Bq m⁻³.

7.1.4 Risks associated with radiation exposure of the population in the Netherlands

The population is not only exposed to decay products of ²²²Rn and ²²⁰Rn, but also to external gamma radiation arising from short-lived radon daughters in soil and building materials. The risk to the Dutch population due to inhalation and external irradiation are evaluated below. The annual mean equilibrium equivalent concentrations of ²²⁰Rn and ²²⁰Rn in the Netherlands were given in Chapter 5 and are summarized in Tables 7.2 and 7.3, as well as the derived average excess mortality risks. Data on the mean annual effective dose equivalent and excess mortality risk from external irradiation due to short-lived daughters are presented in Table 7.4.

Table 7.2. Mean occupancy factors, equilibrium equivalent concentrations and mortality risks from the inhalation of 222Rn and progeny in the Netherlands

Residence area	Occupancy factor	EEC	Exposure rate	Excess mortality risk
		(Bq m ⁻³)	(WLM a ⁻¹)	(10 ⁻⁶ a ⁻¹)
At home: living room	0.37	12	0.06	21
bedroom	0.33	9	0.04	14
Indoors elsewhere	0.18	12	0.03	10
Outdoors	0.12	2	0.003	1
Total	1.00		0.13	46

Table 7.3. Mean occupancy factors, equilibrium equivalent concentrations and mortality risks from the inhalation of ²²⁰Rn and progeny in the Netherlands

Occupancy factor	EEC	Exposure rate	Excess mortality risk
	(Bq m ⁻³)	(WLM a ⁻¹)	$(10^{-6} a^{-1})$
0.37	0.5	0.035	5.3
0.33	0.5	0.031	4.7
0.18	0.5	0.017	2.6
0.12	0.2	0.005	0.7
1.00		0.088	13
	0.37 0.33 0.18 0.12	10.37 (Bq m ⁻³) 0.37 0.5 0.33 0.5 0.18 0.5 0.12 0.2	factor (Bq m ⁻³) (WLM a ⁻¹) 0.37

Table 7.4. Average external irradiation in the Netherlands due to the short-lived radon daughters (*)

Residence area	Occupancy factor	Effective dose equivalent rate (mSv a ⁻¹)	Excess mortality risk (10 ⁻⁶ a ⁻¹)
Indoors Outdoors	0.88 0.12	0.17 0.02	9
Total	1.00	0.19	9

^(*) Including the contribution from 228Ac

The data on ²²²Rn exposure are limited to dwellings and the outdoor environment. Since information about ²²²Rn concentrations in factories, schools and office buildings is lacking, it is assumed that they are the same as the mean concentration in living rooms.

The risk coefficients are commonly given per WLM and, therefore, equilibrium equivalent concentrations, in Bq m⁻³, have been converted to WLM a⁻¹ in Tables 7.2 and 7.3.

According to 7.1.1, a value of 350×10^{-6} WLM⁻¹ seems a realistic estimate for the lifetime lung cancer mortality risk from ²²²Rn. For the current Dutch population of 15 million, the absolute lung cancer mortality due to exposure to ²²²Rn is estimated to be about 700 annually (see Table 7.2; at an exposure rate of 0.13 WLM a⁻¹, the number calculated is $0.13 \times 350 \times 10^{-6} \times 15 \times 10^{6} = \text{about } 700$; a calculation based on the excess mortality risk due to ²²²Rn gives the same value: $46 \times 10^{-6} \times 15 \times 10^{6} = \text{about } 700$).

This estimate can be compared with the evaluation by Jansen et al. (1990), who derived a range of 310-715 per year for lung cancer mortality due to exposure to ²²²Rn and its daughters in the Netherlands. The tables show that the inhalation of ²²²Rn and daughters contributes the most to the excess mortality risk.

The mortality risk from ²²⁰Rn exposure based on dosimetric models (see 7.1.1) is 150x10⁻⁶ WLM⁻¹. According to the data in Table 7.3, the mortality from lung cancer induced by ²²⁰Rn and daughters is estimated to be about 200 per year. The uncertainty in this figure is determined by the uncertainty in the exposure estimate (about a factor of 2; see 10.1.3) and in the risk coefficient (also a factor of 2), and thus is about a factor of 3 (70 - 600 cancer deaths per year). The total mortality from airborne radon is approximately 900 per year (estimated range: 420-2000 per year).

The mortality due to exposure to external gamma radiation arising from short-lived radon daughters in soil and building materials is about 100 per year (see Table 7.4).

The total mortality risk from inhaled radon and daughters and external gamma irradiation due to radon daughters is approximately $70x10^{-6}$ a⁻¹, corresponding to about 1000 cancer deaths annually among the Dutch population (the uncertainty in this value is about a factor of 2, resulting in an uncertainty interval of 500-2000). About 95% of these deaths is attributable to indoor exposure. It should be noted that a possible contribution of leukaemias and mammary carcinomas to the mortality risk from ²²²Rn can increase the total number by several times 10%.

7.1.5 Psychological aspects of radon exposure

The psychological aspects of radon exposure are not supposed to differ significantly from those of exposure to other types of ionizing radiation. Otten and Vlek (1989) considered both direct and indirect perception-induced effects in the measurability of mental impairment. Direct effects of irradiation relate to possible damage in the mentally healthy individual's ability to execute his/her cognitive functions and capabilities. The intensity (absorbed dose) and nature (quality factor) of the radiation are of importance regarding possible impairment to the basic mental functions. At relatively high doses, impairment, primarily in experimental animals, of learning ability, taste aversion, withdrawal symptoms and insensitivity to pain have been observed. In studies on Japanese atomic bomb survivors exposed in utero, Otake et al. (1988) reached the conclusion that of those exposed to 1 Gy at 8-15 weeks after conception, 45% were mentally retarded. The relative risk for exposure during the 8-15 week period is four times greater than that for 16-25 weeks after conception. In addition, Otake et al. (1988) concluded on the basis of intelligence tests among children of 10-11 years of age that exposure at 8-15 weeks after conception resulted in a decrease in IQ of 30 points at a dose of 1 Gy. The possible indirect effects on mental health are primarily associated with the perception of ionizing radiation and, to a much lesser extent, with the nature and intensity of the radiation.

Research into the relation between radiation and perception showed that the use of nuclear energy is assessed as more harmful, more frightening, less controllable and less voluntary than the use of X rays for medical diagnosis. Although the perception of radon exposure has not been investigated, it can be expected that it lies between the perception of the use of nuclear energy and medical applications, since the exposure is not voluntary and only partially controllable. The risk from radon exposure is not known in large sections of the population.

Damage to mental health has been observed among people living in the vicinity of the nuclear power plant near Harrisburg (USA) and those working at the plant after the accident, and in western Germany as a result of the passage of the radioactive cloud originating from the accident in the nuclear reactor at Chernobyl. This damage includes insomnia, overeating, impairment of thinking ability, headache, stomachache, anxiety and depression. Mental disorders (radiation response syndrome) have only been reported for persons who were at a relatively short distance from an accident, atomic bomb survivors, and patients receiving radiotherapy. Among those who are exposed to a radiation source at a greater distance and/or passively, the degree of concern appears to be strongly influenced by economic, political and philosophical factors.

Research into indirect effects of radiation on mental health has so far been limited to nuclear power plants and radioactive waste. Extension to a wider range of radiation sources is therefore important in order to gain more insight into the relation between perception and type of radiation source.

A conscious attempt to find, and possibly make known the houses with high ²²²Rn

concentrations would seem desirable. The arisal of fear must be prevented as much as possible, and any information given during measurements should take this into account.

7.2 RISKS TO AQUATIC AND TERRESTRIAL ORGANISMS

There are no experimental data on the risks to aquatic and terrestrial organisms from exposure to radon. In general, the radiation dose from radon can be attributed entirely to the natural background radiation in the organism's habitat. Plants or animals which consequently run too high a risk will disappear in the course of time through natural selection. Since the radon isotopes are decay products of primordial radionuclides, it can be stated that the existing species of organisms do not run a great risk of extinction as a result of radon exposure. Laboratory studies have been conducted on the radiosensitivity of a very wide range of organisms, involving primarily acute gamma irradiation (Sparrow et al., 1967; Whicker and Schultz, 1982). They show that the acute lethal gamma dose for mammals (2-20 Gy) is, in general, lower than for all other animal and plant species examined.

The annual dose to aquatic and terrestrial organisms estimated in 6.2.1 does not produce any acute effects in mammals. Moreover, in the case of radon, it involves continuous irradiation at a low dose rate, which in general leads to even greater radiation resistence of organisms. Therefore, it does not seem likely that the occurrence of acute radiation effects in aquatic and/or terrestrial organisms due to radon requires consideration.

One of the long-term effects is cancer induction. Studies in laboratory animals have shown that lung cancer can be produced in rats and dogs from inhalation exposures to high ²²²Rn levels. However, with the possible exception of animals living in burrows, the risk of lung cancer seems to be small considering the relatively low doses they receive. Moreover, it is not known whether many animal species live long enough to allow cancers to develop.

7.3 RISKS TO LIVESTOCK

There are no experimental data on the risks to livestock from radon exposure. Respiratory tract tumours have, however, been observed in rats and dogs following inhalation exposure to high ²²²Rn levels and, at extremely high exposures, also acute (non-stochastic) effects, such as radiation pneumonitis.

The estimated exposures of livestock to ²²²Rn in 6.3 are so low that acute effects will not occur. Assuming a risk coefficient for cancer induction in these animals of the same order as that for humans, and taking into account the presumably low exposures in stables and the longer residence time outdoors, the lung cancer risk to livestock resulting from radon will not be greater than the risk to humans. Furthermore, it can also be mentioned here that the life span of livestock is often too short for a cancer to develop.

7.4 SUMMARY AND CONCLUSIONS

Risk estimates for lung cancer resulting from exposure to ²²²Rn and ²²⁰Rn have been catalogued and evaluated. A risk value of 350x10⁻⁶ WLM⁻¹ for lung cancer mortality due to a lifetime exposure to ²²²Rn and progeny was obtained from data on occupationally exposed miners. The mortality risk value derived from data on the Japanese survivors of atomic bombings and irradiated patients, using a quality factor of 20 for alpha radiation and dosimetric models, is consistent with the value for miners. The lung cancer mortality

risk from inhaled ²²⁰Rn and daughters is estimated to be 150x10⁻⁶ WLM⁻¹, based on dosimetric models. The uncertainty in the risk values for ²²²Rn and ²²⁰Rn is about a factor of 2.

The lung cancer mortality resulting from exposure to ²²²Rn and daughters is estimated at approximately 700 per year (uncertainty interval: 350-1400 per year), and that from ²²⁰Rn and daughters at about 200 (70-600) per year. The annual number of cancer deaths from external irradiation due to short-lived decay products of radon in soil and building materials is estimated to be 100 (50-200). There are no indications or data available concerning detrimental effects of radon on ecosystems in terms of threatening extinction of species.

The main conclusions from this chapter are:

- 1. Lung cancer induction is considered to be a risk of radon exposure and death due to radon-induced lung cancer the risk criterion. The relative risk projection model is used for estimating the total risk. Other forms of cancer should also be considered.
- 2. For estimating the lifetime risk of lung cancer mortality due to continuous exposure to ²²²Rn and progeny, an average value of 350x10⁻⁶ WLM⁻¹, or 5x10⁻⁶ Bq⁻¹ m³ a⁻¹ (EEC) can be used. The uncertainty in these values is a factor of 2.
- 3. For estimating the lifetime risk of lung cancer mortality due to continuous exposure to ²²⁰Rn and progeny, an average value of 150x10⁻⁶ WLM⁻¹, or 28x10⁻⁶ Bq⁻¹ m³ a⁻¹ (EEC) can be adopted. The uncertainty is a factor of 2.
- 4. Critical groups have not been sufficiently identified. Possible groups consists of persons occupationally exposed in crawl spaces, cellars or tunnels, and workers at hot springs. Identification of these groups is recommended.

8. MODELLING INDOOR RADON CONCENTRATIONS AND POSSIBLE CONTROL MEASURES

Radon in indoor air originates from various sources, which contribute independently of each other and, depending on circumstances, to a varying degree to indoor concentrations. The principal radon sources are outdoor air, the soil under and near the dwelling*, and building materials. Radiation protection policy has placed these sources in the category "Building construction and occupancy", the objective being risk reduction (VROM, 1991). Other sources of radon in dwellings are radon-bearing natural gas and radon dissolved in tap water (see Table 2.3). Although their contributions are small, they are included in the proposed risk limits. The reader is referred to 5.3 and 5.4.1 for a discussion of the radon exposures resulting from the use of natural gas and drinking water.

In addition to geographical location, building design and building materials, the of the occupants' habits with regard to ventilation is a factor affecting the radon concentration. The three main sources are discussed in more detail in 8.1, followed by the trend in the exposure (8.2), and the effectiveness of radon-control measures (8.3) with, finally, possible scenarios for radon (Section 8.4).

8.1 MATHEMATICAL MODELS

Models consist of the calculation of radon source strengths and air flows inside dwellings, and the solution of systems of differential equations which describe the behaviour of radon concentrations with time. Time-dependent solutions (dynamic models), in which the parameter values vary, are used to increase our scientific understanding of the problem of indoor radon and to verify measurement results (Stoop et al., 1991). Steady-state models use mean parameter values and present the equilibrium situation. These models are employed for analyzing effects of building materials used, building constructions and various types of housing (van den Ham et al., 1990).

8.1.1 Description of a model for calculating radon concentrations

Contribution to radon concentration from outdoor air

Radon concentrations in outdoor air exhibit both daily and seasonal variations. The ventilation in Dutch homes is ordinarily supplied by unfiltered outdoor air which, at typical ventilation rates (0.3-5 h⁻¹), stays for an average of no longer than a few hours inside the dwelling. The decay of ²²²Rn carried with the outdoor air during its transit through the dwelling can be ignored (less than 5%). The outdoor air contribution to the ²²²Rn concentration in the various rooms of the dwelling can be modelled as a simple addition of the outdoor atmospheric ²²²Rn concentration plus the calculated concentrations arising from other sources, independent of the ventilation pattern in the dwelling.

Contribution to radon concentration from building materials

Building materials as a source of radon in the indoor atmosphere differ - for the mathematical model - from infiltration sources such as outdoor air and the soil underneath the dwelling.

(*) For convenience, the word "dwelling" or "house" is also used for other buildings in which people spend a large amount of time, such as schools and offices

The building materials in a room in which one wishes to calculate the radon concentration are taken to be a pure radon source (without daughter products), whereas the radon entering with air is accompanied by an unspecified concentration of short-lived daughters. The radon exhalation rate (or source strength) from building materials is considered to be virtually constant with time, whereas the concentration in the infiltrating air shows large variations.

The contribution to the radon concentration in the room air from the building materials incorporated into the room structure is determined from the radon exhalation rate from the interior surfaces of the room and the air-exchange rate in the room.

For the calculation of the exhalation rate from a wall, only the half-thickness of the inside wall is usually regarded as a source. The ²²²Rn exhalation rate from 1 m² of room surface (see also 1.2.3) is calculated using expression 8.1; the equation for the calculation of the exhalation rate of ²²⁰Rn has the same form.

$$E(^{222}Rn) = (d/2) \rho C(^{226}Ra) \lambda \eta f(D_e)$$
 (8.1)

where $E(^{222}Rn)$ = the exhalation rate of ^{222}Rn (Bq m⁻² s⁻¹) d/2 = the half-thickness of the wall (m) ρ = the density of the building material (kg m⁻³) $C(^{226}Ra)$ = the activity concentration of ^{226}Ra in the building material (Bq kg⁻¹) λ = the decay constant of ^{222}Rn (s⁻¹) η = the radon emanation coefficient $f(D_e)$ = the effective diffusion factor.

This equation is based on the assumption that diffusion is the steering mechanism and that the concentrations on both sides of the wall are approximately equal. However, doubts have arisen about the general validity of this assumption. More work on this within the STRATEGO research programme is recommended.

The factor $d/2 \rho C(^{226}Ra)$ in expression 8.1 indicates the amount of ^{226}Ra in a portion of wall with an area of $1m^2$ and a thickness half that of the wall expressed in activity units (Bq). The numerical value of this factor is also the number of ^{222}Rn atoms generated per second in the specified wall volume. Multiplication by the decay constant of ^{222}Rn yields the activity of ^{222}Rn produced per second. Only a fraction of this, usually less than 30%, enters the air-filled pores of the building material; this fraction is known as the emanation coefficient η . The radon which has made its way into the pores moves through these pores by diffusion and after some time enters the room air; radioactive decay occurs during this time. The fraction reaching the surface of the building material is given by the factor $f(D_e)$. For wall thicknesses of 7-15 cm, this fraction is more than 90% for ^{222}Rn ; because of the short half-life of ^{220}Rn (56 s), the effective exhalation depth is only 0.5 cm.

In some practical cases it is also desirable to add a reduction factor to expression 8.1, which takes into account the exhalation-reducing effect of a finishing coat. A finishing coat usually has a negligible effect on the 222 Rn exhalation rate, but has a large effect on the exhalation rate of 220 Rn because of its short half-life. The contribution [C(Rn), Bq m⁻³] to the radon concentration in the air in a room due to the radioactivity of the building material incorporated into that room is determined by the radon exhalation rate [E(Rn,w), Bq s⁻¹] from the total exhaling surface area, the air-exchange rate $[\lambda_v, s^{-1}]$ in the room, and

by radioactive decay. The process is described by the following balance equation:

$$\frac{dC(Rn)}{dt} = \frac{E(Rn,w)}{V} - C(Rn) \lambda_v - C(Rn) \lambda$$
(8.2)

where V is the volume of the room (m³). The steady-state solution to this equation is:

$$C(Rn,t) = C(Rn,0) e^{-(\lambda_v + \lambda)t} + \frac{E}{(\lambda_v + \lambda)V} (1 - e^{-(\lambda_v + \lambda)t})$$
(8.3)

where C(Rn,0) is the radon concentration in the room air at t=0.

The decay constant λ for ²²²Rn is 0.0076 h⁻¹, much less than the air-exchange rate λ_v (0.3-5 h⁻¹), so that λ can be ignored in this equation. The value of λ for ²²⁰Rn is 44.6 h⁻¹, so that in this case λ_v can be ignored and the ²²⁰Rn concentration in indoor air is almost independent of the air-exchange rate. It follows from expression 8.3 that, for a constant λ and E(Rn,w), the contribution to the radon concentration from building materials eventually reaches the value:

$$C(^{222}Rn)_b = \frac{E(^{222}Rn,w)}{\lambda_v V}$$
 (8.4)

It furthermore shows that after disturbing the steady-state situation, a new equilibrium is established within a few hours; for example, at an air-exchange rate greater than 0.8 h⁻¹, more than 80% of the equilibrium value is attained in less than two hours.

The influence of building materials used in other rooms is only important when there is airflow from these rooms to the room under study. If such is the case, these air flows must be inserted into the activity balance equation (expression 8.2) as source terms. This source term has the form:

$$S_{i} = \underline{qi} C_{i}(Rn)$$
 (8.5)

where S_i = the source strength of Rn carried with air from room i (Bq m⁻³ s⁻¹) q_i = the airflow rate from room i (m³ s⁻¹) $C_i(Rn)$ = the radon concentration in room i

It is implicitly assumed that the radon gas entering with infiltrating air or from exhalation is distributed uniformly throughout the volume of the room almost instantly.

For systems of several rooms connected by airflows, all activity balance equations (one for each room) must be solved simultaneously (Ackers, 1986). The KVI of the University of Groningen has worked out solutions to the type of equations of expression 8.2 for the dynamic model.

The essence is that λ_v has been made time-dependent by calculating the air flows from variations in atmospheric pressure differences in and around the dwelling (Stoop et al., 1991).

The airflows follow from the empirical equation:

$$q_{ik} = T_{ik} \qquad \left[\begin{array}{c} \frac{\Delta P_{ik}}{1 \text{ Pa}} \end{array} \right] 1/n \tag{8.6}$$

where qik

 q_{ik} = the airflow rate from room i to room k (m³ s⁻¹)

 ΔP_{ik} = the atmospheric pressure difference between rooms i and k (Pa)

 T_{ik} = air leakage between rooms i and k at a pressure difference of 1 Pa $(m^3 s^{-1})$

n = an exponent which determines the nature of the air flow $(1 \le n \le 2)$; n = 1, laminar flow; n = 2, total turbulent flow

The air-exchange rate results from the total of all airflows to a room and the volume of the room.

By means of step-by-step calculation, with λ_v being held constant for a short time, the buildup of the radon concentration is calculated from the result of the successive steps.

Radon contribution from the soil beneath the dwelling

The radon flux from the soil surface (or source strength) is not constant and is governed by two mechanisms: diffusion of radon through the soil pores, and pressure-driven flow of radon-bearing air through soil. The first process is affected by changes in moisture content and water table; the second process depends on the pressure distribution in and around the dwelling, caused by temperature differences and winds.

Once the radon flux from the soil immediately under the dwelling (for example, into the crawl space) has been found by means of calculations on a physical model, its contribution to the radon concentration inside the dwelling can be calculated, employing the same method as that used for the contribution from building materials. The diffusive flux from the soil is treated in the same manner as the exhalation rate from the building material, and the pressure-driven flow of air through the soil analogous to airflows from other rooms.

Much work is still required on refining the modelling of the flux from the soil. Furthermore, the approach is dependent on the current building construction practices. In the USA, where houses are commonly built with a basement substructure of which the wall surface below the soil grade is in direct contact with the soil, other processes prevail than in the Netherlands, where most of the houses built have a crawl space.

Diffusive transport in soil

The ²²⁶Ra content of the upper 1 to 2 m of soil determines the production rate of radon in the soil. The fraction of the radon formed in the solid grains which enter the pore volume of the soil (emanation coefficient) can then migrate through the pore system by diffusion to be exhaled at the surface (see also 1.1.2 and 1.2.3).

The nature and size of the soil pores are described by two factors: the porosity of the soil and the tortuosity of the pores. The porosity, ϵ , is defined as the ratio of the pore volume to the total volume of the soil.

The tortuosity of the pores, λ , can be defined as the ratio of the distance between two points in the soil to the shortest path length between these two points via the pores.

Because the moisture content of the soil affects the diffusion coefficient and the emanation coefficient (1.2.3), mathematical models distinguish at least three layers of soil. The first

is a water-saturated layer (groundwater), the second is a layer with only air-filled pores, and the third is a layer in which the grains are covered with a thin moist film (van den Ham et al., 1990). In the last layer, the tortuosity factor should actually be reduced slightly because the diffusion path may become partly blocked. Because of other uncertainties, however, this refinement is seldom built into the models.

The diffusion length of radon is much greater in air-filled pores (approximately 1 m) than in water-filled pores (approximately 1 cm) (Tanner, 1990; Fleischer, 1987).

Each of the modelled soil layers has its own fixed value for the parameters controlling the diffusion process. By placing boundary conditions on the interfaces between the layers, the different diffusion equations are linked, so that the constants included in the mathematical solution can be validated.

The system of diffusion equations can be described in a one-dimensional or a multidimensional form. An example of a one-dimensional equation for a layer is given in expression 8.7:

$$\frac{\delta C}{\delta t} \approx D_e \frac{\delta^2 C}{\delta x^2} - \lambda C + P \tag{8.7}$$

where C = the activity concentration of radon per unit volume of pore space at depth x below the surface and time t (Bq m⁻³)

 λ = the decay constant for radon (s⁻¹)

 D_e = the effective diffusion coefficient of radon in the soil pores (m² s⁻¹)

P = production rate of radon per unit volume of pore space (Bq m⁻³ s⁻¹).

The effective diffusion coefficient, D_e , is obtained from the diffusion constant, D_m , which applies to diffusion of radon in a large volume of the fluid in the pores, using the equation:

$$D_{e} = D_{m} \nu \tag{8.8}$$

The production rate, P, is obtained from:

$$P = C(Ra) \rho \eta \lambda / \epsilon$$
 (8.9)

The steady-state solution, i.e. f(dC,dt) = 0, can be obtained as:

$$C(x) = A e^{\alpha x} + B e^{-\alpha x} + s \tag{8.10}$$

where
$$\alpha = (\lambda/D_c)^{1/2}$$
 and $s = P/\lambda$ (8.11)

The factors A and B are determined from boundary conditions. These are:

- a. the radon concentration in the pore volume at the interface between two layers should give the same result for both layers;
- b. at the interface, the net radon flux from a layer should equal the net flux into the adjacent layer;
- c. at the ground surface, the concentration in the pore gas should equal that in the air above it:
- d. at great soil depths, the gradient of the radon concentration is zero and the radon

concentration is finite.

The diffusive flux from the soil into the dwelling, J_o (Bq m⁻² s⁻¹), is given by:

$$J_o = -\epsilon D_e \frac{dC}{Dx}$$
 | ground surface. (8.12)

Flow of air through the soil

Airflows (ventilation) in a dwelling arise from pressure differences between indoors and outdoors and between rooms (floors). These pressure differences, commonly of the order of a few Pascals, are caused by wind and temperature differences between indoors and outdoors; they can be calculated from ventilation models (De Gids and Pfaff, 1986).

In general there will be a pressure difference between the crawl space and outdoors, which drives the ventilation of the crawl space. The same pressure difference also drives a small flow of air through the soil; from outdoors through the soil into a structure on the windward side and from indoors through the soil to outdoors on the leeward side.

The air velocity, ν (m s⁻¹), through soil is, in addition to the pressure gradient, dP/dx, controlled by the (air) permeability, k (m²), of the soil through the expression (see also 1.1.3)

$$\nu = - \underline{k} \cdot \underline{dP}$$

$$\mu \quad dx \tag{8.13}$$

where μ is the viscosity of air: 1.8x10⁻⁵ kg m⁻¹ s⁻¹.

At a given pressure difference between crawl space and outdoors, the pressure gradient in soil is also determined by the length of the path through the soil; this depends, for example, on the depth and the shape of the foundation of the dwelling, since the airflow will pass under the structurally intact part of the foundation.

Under normal conditions, the air velocity through the soil to the crawl space is less than $1x10^{-4}$ m s⁻¹ and the total flow rate less than $1 \text{ m}^3 \text{ h}^{-1}$. The residence time in the soil during transport is therefore so long, a few hours, that much radon is taken up into this air: the radon concentration in the air after its movement through the soil is commonly of the order of a few kBq m⁻³. As a result, despite the low flow rate, the contribution to the radon concentration inside the dwelling is often substantial.

8.1.2 Calculation of radon decay-product concentrations

Calculation of the concentrations of radon daughters in air is important because the radiation dose from inhaled daughters is large compared with the dose from radon itself (see 6.1.4).

To calculate the respective radon daughter concentrations, activity balance equations, one for each daughter, must be formulated and simultaneously solved (Ackers, 1986).

In addition to generation by the decay of airborne radon and removal by radioactive decay, and supply and removal by ventilation, the removal of the daughters from the air by deposition on the walls, floor and ceiling, and furniture in the room is taken into account (plateout effect). Since the plateout depends strongly on whether or not the daughters are attached to aerosols, both the attached and unattached fractions are

calculated for all short-lived radon daughters.

The plateout rate is commonly described using removal constants which, mathematically speaking, play the same role as the decay constants. This is also true for the rate of attachment to aerosols, the constants of which are a function of the aerosol concentration in the room air being examined. Allowance is also made for the recoil effect: upon alpha decay of attached nuclides, the decay product created may become detached from particles as the result of its high recoil energy. The probability of recoil detachment from aerosol particles upon beta decay is considered to be zero.

A few results of daughter concentrations, calculated for this complex process, are included in the next section.

8.1.3 Examples of results of model calculations

A computer programme has been developed specifically for the Dutch situation in "Building construction and occupancy" (Ackers, 1986). The model has been used to determine the limits to be set on the radioactivity concentrations in building materials (van Heijningen and Ackers, 1990).

The calculated radon daughter concentrations are compiled in Table 8.1 for a typical single-family dwelling. In this case the radon concentration in room 2 is higher than in room 1 because room 2 is ventilated with air originating from room 1 and from the crawl space. With the emphasis on the soil as a radon source for the dwelling, a mathematical model has been constructed containing soil, crawl space and dwelling, which limits itself to calculating the radon source strength and the resulting radon concentrations in the steady state. It has been used in an exploratory study for evaluation of constructional measures (van den Ham et al., 1990).

A few relevant results for a house with a crawl space are summarized in Table 8.2.

Experience gained with calculations for different types of houses indicates that building materials contribute about 20 Bq m⁻³ at most to the indoor ²²²Rn concentration (average about 8 Bq m⁻³), while the maximum contribution from outdoor air is about 10 Bq m⁻³ (average about 3 Bq m⁻³) (see also 5.4).

The contribution from soil shows the largest variation; from the measured maximum ²²²Rn concentration (three-month average) in Dutch dwellings of 120 Bq m⁻³ it follows that the contribution from soil can be as much as about 100 Bq m⁻³.

The ²²⁰Rn concentration due to building materials is estimated to average 2 Bq m⁻³. Taking an equilibrium factor of 0.4 for ²²²Rn and of 0.1 for ²²⁰Rn in indoor air, dose conversion coefficients of 0.14 mSv a⁻¹ and 0.6 mSv a⁻¹ per Bq m⁻³ (EEC) for ²²²Rn and ²²⁰Rn respectively (see Table 6.5), and an indoor occupancy factor of 0.88, the average annual dose caused by ²²²Rn and ²²⁰Rn exhaled from the building materials is found to be approximately 0.4 mSv and 0.1 mSv, respectively. The external radiation dose from shortlived radon daughters present in the building materials is about 0.2 mSv a⁻¹ on average (see 7.1.4).

Table 8.1. Calculated radon and radon daughter concentrations in two rooms in a dwelling, room 1 being ventilated with outdoor air and room 2 with air from other rooms in the dwelling (concentrations in Bq m³; ventilation rate in h¹) (Ackers, 1986)

Parameter	Outdoors	Room 1	Room 2	
²²² Rn conc.	4.0	11.2	26.1	
218Po conc., free	0.4	1.5	3.4	
²¹⁸ Po conc., att.	2.6	6.2	14.7	
214Pb conc., free	0.0	0.1	0.27	
214Pb conc., att.	3.0	4.5	10.4	
²¹⁴ Bi conc., free	0.0	0.002	0.006	
²¹⁴ Bi conc., att.	3.0	4.0	8.9	
F (²²² Rn)	0.75	0.42	0.41	
EEC (222Rn)	3.0	4.7	10.7	
²²⁰ Rn conc.	4.0	4.0	4.1	
216Po conc., free	4.0	4.0	4.1	
²¹⁶ Po conc., att.	0.0	0.015	0.015	
²¹² Pb conc., free	0.0	0.003	0.003	
²¹² Pb conc., att.	0.1	0.45	1.0	
212Bi conc., free	0.0	0.0	0.0	
²¹² Bi conc., att.	0.1	0.28	0.71	
F (²²⁰ Rn)	0.03	0.11	0.24	
EEC (220Rn)	0.12	0.43	0.98	
Ventilation rate	na	0.95	0.50	

(na) not applicable

The fraction attached to aerosols is indicated with "att".

Attachment rate to aerosols is 60 h^{-1} . Plateout rate of the free fraction is 20 h^{-1} . Plateout rate of the attached fraction is 0.2 h^{-1} .

Both rooms receive a small supply of air from the crawl space. Radon concentration in the crawl space is 42 Bq m⁻³

Table 8.2. A few parameters and calculated results for a dwelling with a crawl space, using a model which takes account of diffusive transport from and pressure-driven airflows through the soil

Parameter	Value	Unit
222Rn exhalation rate from building material	3.0	Bq m ⁻³ h ⁻¹
Air temperature indoors	20	°c̄
Air temperature outdoor	10	°C
Wind speed	2	m s ⁻¹
²²² Rn concentration in outdoor air	4	Bq m ⁻³
²²² Rn concentration in the soil	20	Bq m ⁻³
Average radon emanation coefficient	0.15	•
Density of the soil	1700	kg m ^{.3}
Porosity	0.35	
Tortuosity	0.7	_
Permeability	1 · 10 ⁻¹¹	m^2
Airflow through soil	0.03	$m^3 h^{-1}$
Ventilation rate in dwelling	0.8	h ⁻¹
Ventilation rate in crawl space	0.2	h ⁻¹
²²² Rn exhalation rate from soil	15	Bq m ⁻² h ⁻¹
²²² Rn concentration in air flow through soil	6300	Bq m ⁻³
²²² Rn concentration in dwelling	20	Bq m ⁻³
²²² Rn concentration in crawl space	160	Bq m ⁻³
Contribution to 222Rn concentration in dwelling		_
from outdoor air	4	Bq m ⁻³
from building material	4	Bq m ⁻³
through floor slab	12	Bq m ⁻³
Contribution to 222Rn concentration in crawl sp	ace	_
from outdoor air	4	Bq m ⁻³
from building material	22	Bq m ⁻³
diffusive transport from soil	110	Bq m ⁻³
airflow through soil	24	Bq m ⁻³

8.1.4 <u>Calculation of the dose contribution from building materials (limit-value expression)</u>
For the purpose of controlling the risks resulting from "Building construction and occupancy" (VROM, 1991), the limitation method has been developed for the risks due to the use of (substitute) building materials (van Heijningen and Ackers, 1990). This method is briefly outlined here, and has been used in the scenarios in 8.4.

The annual effective dose equivalent (D) resulting from naturally occurring radioactive nuclides in building materials consists of three components:

$$D = D(^{222}Rn) + D(^{220}Rn) + D(gamma)$$
 (8.14)

The D(²²²Rn) and D(²²⁰Rn) dose contributions resulting from the inhalation of radon decay products can be calculated using the models described in 8.1.1; the D(gamma) contribution from external irradiation is calculated from the gamma radiation intensity from the decay of radionuclides in the ²²⁶Ra and ²³²Th series and of ⁴⁰K present in building materials. The exposure rate from gamma radiation in a room depends on the type of building materials, their dimensions and masses, and is independent of the ventilation pattern, temperature or humidity in the dwelling.

The dose contribution of a building material (D_m) in a dwelling can be calculated separately from the other building materials (D_r) . D_m can be expressed in activity concentrations as (van Heijningen and Ackers, 1990):

$$D_{m} = [(a + b \eta)C_{Ra} + (c + d \eta)C_{Tb} + e C_{K}] \rho 10^{-6}$$
(8.15)

where C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq kg⁻¹) of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in the building material being considered and

 η = the radon emanation coefficient (assumption: the coefficients for 222 Rn and 220 Rn are the same)

 ρ = the density of the building material (kg m⁻³)

a,b,c,d,e, = constants

The annual effective dose equivalent from all building materials together can be limited:

$$D = D_m + D_r < D_{timit}$$
 (8.16)

In order to obtain a "standstill" of the doses due to radon and gamma radiation from building materials, dose limits have been set for the five most commonly used materials (see Table 8.3), which together constitute about 90% of the current Dutch market. This has been accomplished by setting the highest value (D_{max}) for D_{limit} which, for the material being considered, is calculated for D when the maximum value (for common material) is inserted for the various parameters. The limits have been calculated assuming maximum use of the material in a standard house. The parameter values obtained for apartments are summarized in Table 8.3.

Most of the currently available building materials are well below the limitation criterion D_{max} . From a radiation protection point of view, it is undesirable that the difference is

made up by using other raw materials. Should this occur in practice, revising the current limits downwards could be considered.

Table 8.3. Parameter values for the limitation expressions [see (8.15) and (8.16)] of Dutch building materials used in flats (van Heijningen and Ackers, 1990)

Parameter	Concrete	Brick	Sand-lime brick	Aerated concrete	Gypsum products
a	1.61	0.53	0.51	1.16	0.82
b	34.60	13.98	13.97	21.99	16.74
C	1.98	0.61	0.60	1.35	0.96
d	11.80	6.73	6.73	9.49	9.50
е	0.16	0.05	0.05	0.10	0.07
D, (mSv a ⁻¹)	0.04	0.64	0.64	0.53	0.52
D _{max} (mSv a ⁻¹)	1.36	0.98	0.76	0.71	0.64

8.2 THE TREND IN THE EXPOSURE LEVEL

In Section 5.5 a calculation was made of the average exposure levels to ²²⁰Rn and ²²²Rn in the Netherlands. It is important to know, particularly from a policy point of view, whether this level can be expected to change in time. In their survey, Meijer et al. (1986) examined whether a relationship existed between the year of construction of the dwelling and the indoor ²²²Rn concentration. They found that the concentration in houses built after 1970 was about 5-10 Bq m⁻³ greater than in houses built before that date. A possible explanation for this is the larger proportion of concrete now used in building construction. In view of the relatively high ²²²Rn exhalation rate from this material (see 5.2), a slight increase in the concentration can be expected. A possible contributory cause which should be mentioned is that the more air-tight outside wall - hence lower ventilation capacity - of present-day homes, which can cause higher concentrations.

It is hard to predict to what extent the observed upward trend in the indoor ²²²Rn concentrations will continue in the coming years. As has been explained in the preceding sections, the concentration is the resultant of a large number of processes. Changes in these processes can lead to higher as well as lower concentrations. If it is assumed that the increased use of concrete and the reduction in ventilation rates are indeed the causes of the observed increase, then the situation will stabilize around the year 2020 because of the limits set on them. Ultimately, the mean ²²²Rn concentration will be greater than the current value of 29 Bq m⁻³ in living rooms. The above data and data on the number of dwellings built after 1970 indicate that, conditions being equal (including construction practices, building materials and ventilation rates), the mean value will have increased by about 5 Bq m⁻³, or about 15%, by the year 2020.

8.3 POSSIBLE RADON CONTROL MEASURES

8.3.1 Survey of possible measures

There are a large number of possibilities for reducing indoor levels of ²²²Rn and ²²⁰Rn. They fall into three categories:

- measures which directly reduce the source strength
- measures which seal off the source or otherwise reduce it

- measures which directly influence the airborne radon concentration.

Possible measures, specifically related to the Dutch situation, are given by Fischer (1989) and van den Ham et al. (1990), including as the most important:

- a) Limiting ²²⁶Ra concentration in the building materials.
- b) Drawing off the soil gas by means of:
 - 1. Covering the soil in unpaved crawl spaces with plastic sheeting or concrete, and then drawing soil gas off at several points from beneath the barrier thus created with a fan.
 - 2. Mechanical ventilation via a drainage system built around the dwelling.
- c) Covering the soil in unpaved crawl spaces with:
 - 1. Plastic sheeting.
 - 2. Concrete.
- d) Increasing the (natural) ventilation rate in the crawl space by enlarging the vents or using mechanical ventilation.
- e) Improving the airtightness of the floor slab by:
 - 1. Sealing the joints and openings around service entrances.
 - 2. Applying a coating above or below the floor slab.
- f) Increasing the natural ventilation in the dwelling by enlarging the vents.
- g) Combining the measures mentioned under d) and e)1.

It should be pointed out that almost no fieldtesting of the above measures has as yet been done in the Netherlands, so that this is a preliminary survey only.

8.3.2 Effectiveness of measures

A number of methods for determining the effectiveness of a control measure are in common use. In the first method, the houses (of the same type) to be studied are divided into two groups. The measure is implemented in the first group while the second group serves as a control.

Since the radon concentration varies substantially among the houses within each group, the size of the groups has to meet minimum requirements which are related to the effectiveness of the measure and the desired reliability. This method has been employed by Put (1991) among others.

The second method compares the situation before and after remedial action in the same dwelling. Since the radon concentration varies with time (see 5.4.1), a comparison cannot always be easily made. This method has been applied in the Dutch test house in Roden (de Meijer, 1991) and by Swedjemark and Mäkitalo (1990) in Sweden.

A third method is to conduct laboratory-scale tests of the measure under controlled conditions. An observed effect can then be directly attributed to a particular alteration made. Although this possibly does not paint a correct picture of the absolute effectiveness of the measure in practical situations, it does establish the order of relative effectiveness (see, for example, van Dijk and de Jong, 1989). Finally, the effectiveness of measures can be assessed by using models which describe the radon concentration in a dwelling (see 8.1). This will be discussed at the end of this section.

Relatively little research into the effectiveness of the different measures has as yet been carried out in the Netherlands. However, several studies have been conducted abroad, for example, by Swedjemark and Mäkitalo (1990) in Sweden. Their study revealed that if the

soil is the principal source of indoor ²²²Rn, drawing soil air from the ground beneath the dwelling ("sub-slab suction") or installing "radon wells" around the house should be regarded as the most effective control techniques. The latter measure offers the radon a faster escape route, leading to lower concentrations in soil gas and thus reduced infiltration of radon into the dwelling. When the building materials are found to be the major source, the ventilation rate in the dwelling has to be increased. This study assessed the effect of increasing the natural ventilation rate in the crawl space by enlarging the vents (65% reduction in radon concentration) to be inadequate. Mechanical ventilation of the crawl space reduced the radon concentration by an average of 85% (Swedjemark and Mäkitalo, 1990).

In the United States measures focus on the infiltration of radon-bearing air from soil into indoor air. The measures mostly involve sealing the walls and floors in direct contact with the soil such as, for example, a basement substructure. In addition, studies are devoted to (usually mechanical) exhaust ventilation of the soil (EPA, 1987). Attention has also been paid to reducing the ²²²Rn daughter concentration. According to Moeller and Fujimoto (1984), the installation of six unipolar ion generators in the ventilation system achieved a fourfold reduction in the daughter concentration. For several reasons, caution is required in translating the results obtained abroad for the Dutch situation. Firstly, the investigations are usually conducted in dwellings with much higher ²²²Rn levels than are observed in the Netherlands (200-4000 Bq m⁻³). In addition, the difference in building characteristics (e.g., cavity wall, crawl space) hamper direct comparison. The best approach to validating measures is implementing them in a Dutch practical situation and using Dutch materials. As previously mentioned, little attention has as yet been given to this subject. A few Dutch studies are discussed below.

Stoop et al. (1991) conducted experiments in a test house on mechanical exhaust ventilation of the crawl space, and succeeded in halving the ²²²Rn concentration in the living room. The same technique was applied in Arnhem in a dwelling with a high ²²²Rn level (Put, 1991). The effect of this measure is presented in Table 8.4. It is seen that the concentration in the living room of this house was also reduced. The exhaust ventilation system lowers the pressure in the crawl space in relation to the living room, thereby preventing radon infiltration.

Table 8.4. Annual average ²²²Rn concentration (Bq m⁻³) before and after the installation of a mechanical ventilation system in the crawl space (Put, 1991)

mechanical ventilation	mechanical ventilation
325 - 480	260
116 - 158	32
40	23
50	21
	116 - 158 40

The effect of a barrier between the crawl space and the underlying soil was examined in a row of identical houses (Put, 1991). Two houses had unpaved crawl spaces; in two other houses the crawl space soil had been covered with aerated concrete and plastic sheeting and in one house with aerated concrete only. The measured ²²²Rn concentrations in the

living rooms averaged 39, 36 and 32 Bq m⁻³, respectively. Although the concentration appears to be reduced, it is probably not by very much. However, the number of houses studied was too small.

The water table significantly influences the ²²²Rn concentration in soil gas (Loos et al., 1991). As the water table falls, the ²²²Rn concentration in soil gas increases, and vice versa. After a perimeter drainage system was installed to go around the test house in Roden, the water table fell by about 50 cm. However, the anticipated increase in the concentration in the crawl space did not occur. It is supposed that the frequent opening of the crawl space for carrying out experiments is one of the causes. Another cause is probably that it is not the water table which is the determinant of the radon flux from the ground but the level of the water-saturated layer, of which the top was still near the soil surface of the crawl space after the drainage system had been installed.

An investigation of the influence of paints used in the Netherlands on the ²²²Rn exhalation rate from building materials shows that only paints based on chlorinated rubber, epoxy resin and polyurethane produce a significant reduction (van Dijk and de Jong, 1989). These paints decreased the ²²²Rn exhalation rate by 20-95%. The reduction depends on the type of paint, the number of coats applied and on whether the paint is applied to one or both sides of the building material. The paints and techniques ordinarily used in the Netherlands have only a marginal effect on radon exhalation.

Owing to the lack of experimental results in the Netherlands, the effectiveness of the measures mentioned in 8.3.1 has been calculated using a mathematical model developed by TNO (van den Ham et al., 1990; see also 8.1). The calculations are based on an average and an unfavourable situation. The associated characteristic parameter values are:

Parameter	Unit	Average situation	Unfavourable situation
222Rn concentration in the house	Bq m ⁻³	20	60
²² Rn concentration in the crawl space Radiation dose from residence in houses	Bq m ⁻³ mSv a ⁻¹	180 1.2	840 3.2

The results are presented in Table 8.5. The mathematical model found that most measures produce a similar effect.

8.3.3 Cost effectiveness of measures

Using the results reported in Table 8.5, van den Ham et al. (1990) made an estimate of the cost effectiveness of the different control measures. By this is meant the ratio of the dose reduction to the total annual cost of a measure, expressed in mSv kf⁻¹ (mSv per 1000 Dutch guilders). In addition to the purchase and installation costs of a control technique, the costs of maintenance and energy have also been taken into account. Because of the uncertainties in the above-mentioned model calculations, the cost-effectiveness estimates have been categorized into three groups. The results of the calculations are presented in Table 8.6, for both future and existing houses. They show that for new construction, the cost effectiveness of various control measures is moderate or very good. The use of concrete having a low ²²⁶Ra activity concentration, in combination with the application of one of the other measures, could lead to a dose reduction of about 0.4 mSv per year in an average situation, at an estimated annual cost of Dfl.150.-to Dfl.250.- per house.

Table 8.5. Reduction in the indoor 222Rn concentration after applying a particular measure in new dwellings

Meas	ure Reducti	on in 22	² Rn c	concentration
		ituatio Bq m ^{.3})		nfavourable situation (Bq m ^{.3})
a)	Concrete with low 226Ra concentration	1 -	3	2 - 3
b1)	Exhaust ventilation of the soil	5 ~	9	20 - 40
b2)	Exterior drainage system with mechanical ventilation	2 -	6	10 - 20
c1)	Cover soil in crawl space with plastic sheeting	2 -	6	10 - 30
c2)	Cover soil in crawl space with concrete	2 -	6	10 - 30
d)	Increase the natural ventilation in crawl space	2 -		15 - 40
e1)	Floor slab, seal joints	2 -	6	5 - 25
e2)	Floor slab, PUR coating	3 -	7	10 - 30
f)	Larger vents *	1 -	6	-5 - 20
g)	d) + e1)	6 -	12	25 - 45

^{*} Concentration may be increased

Table 8.6. Categorization of measures on the basis of cost effectiveness, based on results from model calculations; almost no fieldtesting has as yet been carried out.

Mea	feasure		Average situation		Unfavourable situation	
		Future	Existing	Future	Existing	
a)	Concrete with low 226Ra concentration	В	- .	В		
b1)	Exhaust ventilation of the soil	A	A	В	В	
b2)	Exterior drainage system with mechanical ventilation	A	A	В	A	
cl)	Cover soil in crawl space with plastic sheeting	A	A	В	В	
c2)	Cover soil in crawl space with concrete	e A	A	В	В	
d)	Increase the natural ventilation rate in crawl space	В	A	С	В	
e1)	Floor slab, seal joints	В	A	С	В	
e2)	Floor slab, PUR coating	A	A	В	В	
f)	Larger vents	A*	A*	B*	A*	
g)	d) + e1)	В	A	C	В	

category A: not cost-effective (0 - 2 mSv kf⁻¹)

category B: moderately cost-effective (2 - 10 mSv kf-1)

category C: very cost-effective (> 10 mSv kf^{-1})

(*) The possibility of a negative effect is not negligible (-) not applicable

In the case of an unfavourable situation in new construction, application of the d) + e1) combination is expected to be adequate in most instances for reducing the radon exposure to the average level (see Table 8.5).

8.4 SCENARIOS

Possible future developments in the individual and collective doses from indoor inhalation of radon are discussed in this section. The scenarios considered here to break the trend outlined in 8.2 can be divided into those pursuing or not pursuing an active policy. Active policy makes a distinction between radon-control measures in future and existing housing. Measures can be directly implemented in houses during construction at a much lower cost than in existing houses. Current policy is aimed at formulating reduction objectives for radon in dwellings (VROM, 1991).

An exploratory study of the possible effectiveness of radon-reduction measures has been carried out in the Netherlands (see 8.3). No research has as yet been conducted into the possible impact of any measures taken on the radiation dose in the longer term. This section is therefore limited to a primarily qualitative description of a number of possible scenarios.

8.4.1 No control measures

The ²²²Rn concentrations in Dutch homes are generally well within the guidelines recommended by the EC (see Table 1.4). Therefore, a possible scenario is to take no active measures to reduce the concentrations in indoor air. The implication of this approach is that the increase in the average concentration outlined in Section 8.2 will occur. If the current trend were to be extrapolated to the future, then according to the available data the ²²²Rn levels would increase by about 15%. At the moment, however, a number of independent developments can be expected, which will also directly or indirectly influence the magnitude of the ²²²Rn concentration. A few examples are:

- the new Building Decree, which specifies stricter criteria for the air-tightness of the floor slab, so reducing the infiltration rate of radon-bearing soil gas;
- the exhaustion of the gravel reserves in the Netherlands implies that alternatives have to be used in the future, with possible consequences for the radon exhalation rate from concrete:
- shifts in construction practices and/or in use of certain building materials (for example, towards gypsum) can also cause a change in concentrations.

Quantification of the net effect of the independent developments is at this stage not possible.

8.4.2 Control measures in future houses

For new houses, three scenarios can be envisaged which lead to a reduction in the average radiation dose resulting from "Building construction and occupancy":

- reduction of the radiation dose from building materials;
- reduction of the radon infiltration from soil;
- a combination of these two measures.

A reduction of the radiation dose from building materials can be effected through the limit-value expression described in 8.1.4 by choosing a lower value for D_{max} . Clearly, this will have implications for some types of building materials commonly used in the construction of Dutch houses. The extent to which D_{max} can be lowered without appreciably affecting the current construction practices is not known. Nor have the possible financial consequences of such an approach been examined. Because most of the present building materials lie well within the limits (and will remain so), the reduction in the individual dose rate, averaged over all future houses, will only be small. Estimations show no more than 0.1 mSv a^{-1} , on average, for occupants of these dwellings.

Since the soil is usually the major cause of the relatively high radon levels observed in some houses, attention should primarily be focused on reducing the infiltration rate from

the soil. Increasing the airtightness of the floor slab can, if necessary, be combined with increasing the ventilation rate in the crawl space (see 8.3.2 and 8.3.3; Engels, 1989). The question remaining is what desirable level/limit should be adopted for the annual average ²²²Rn concentration in new houses. According to Engels (1989), a standard of 10 Bq m⁻³ should be attainable. Such a concentration implies that the floor slab must be completely sealed. Model calculations have shown that an average 222Rn concentration reduction of about 8 Bq m⁻³ could be achieved in new homes (see 8.3.3). Using the conversion coefficient for ²²²Rn given in 6.1.5, this corresponds to a reduction in individual dose of 0.4 mSv a⁻¹.

The combination of limiting the radioactivity in building materials and reducing the radon infiltration rate can yield a dose reduction of 0.5 mSv a-1 per occupant. Assuming an average of 2.6 persons per household and an annual output of 100,000 new houses, the reduction in the collective dose can be estimated to be 130 person-Sv per year. This corresponds to an annual reduction of 0.6% in the average ²²²Rn exposure of the population in the Netherlands.

8.4.3 Control measures in existing houses

An investigation of the ²²²Rn levels in 900 Dutch homes (de Meijer et al., 1986) found that the percentage of the housing stock exceeding a specified concentration is approximately distributed as indicated in Table 8.7 (see also Figure 5.2). Assuming a total of about 6 million housing units in the Netherlands and taking a remedial action level of 50 Bq m⁻³, the number of houses requiring remediation would be 480,000. The measures will (ultimately) decrease the annual average 222Rn concentration in the houses in question decreases by 15-30 Bq m⁻³ on average (see Table 8.7), corresponding to an individual dose reduction of 0.8-1.5 mSv a-1. Table 8.7 also shows the collective dose reduction which such remedial action would yield. Clearly, the lower the action level set, the greater the possible reduction will be, and likewise the costs involved.

Table 8.7.	Percentage of	the housing stock with 222Rn concentrations
	exceeding the	indicated value and their average concentration
	(de Meijer et	al., 1986)

²²² Rn concentration		Percentage of the housing stock	Number of houses (***)	Average ²²² Rn concentration (*)	Collective dose reduction (**)
В	d w ₋₃	8	,	Bq m ⁻³	8
-	25	50	3,000,000	40	29
>	50	8	480,000	70	5
>	60	5	300,000	80	3
>	75	1.3	78,000	105	1.3
>	100	0.7	42,000	120	0.5
>	200	~ 0.1	6,000		
>	400	~ 0.01	600		

Should remedial action be considered, the problem then arises that no one is familiar with the houses involved. By combining (partly) available information, it might be possible to considerably limit the number of locations and in this way identify risk areas. The

Estimated from the original data (rounded-off figures)
Remedial action if the indicated 222Rn concentration is exceeded

^(***) Assumption: the full housing stock totalled 6,000,000 units as of 1 September 1991

following available information can be important:

- data on the location- and housing type-dependent ²²²Rn concentration in indoor air;
- data on outdoor ²²²Rn concentrations;
- geological information pertaining to the Netherlands;
- data on water tables;
- regional exposure-rate results.

At present, detailed data on the regional differences in exhalation rates from soil are not available. In his publication, Engels (1989) advocated that phosphogypsum blocks also be included in the considerations as a risk-enhancing factor. However, data on important factors, such as the airtightness of the floor slab and the average ventilation rate in the crawl space, are not available.

An investigation of the ²²²Rn concentrations in the remaining houses in the risk areas, for which a rapid (inexpensive) screening method is required, can be conducted. After measurement criteria still to be formulated are exceeded, the radon concentration has to be verified using a track etch detector in a cup before implementation of a control measure is undertaken. This measurement also serves as a basis for determining the effectiveness of the control technique. Next, criteria must be formulated or developed in practice to establish the most suitable measures for a particular housing type.

8.5 SUMMARY AND CONCLUSIONS

The radon (daughter) concentrations in dwellings can be calculated using physical models based on radon source strengths, entry routes, ventilation patterns and aerosol data. In addition to diffusive transport, pressure-driven flow of air through the soil is also included in the model. The uncertainty in the parameters controlling the indoor radon concentration is great. Also, there is still too much uncertainty about whether all entry paths into the dwelling are known. So far, verification of the calculated radon concentrations against values measured in practical situations has been insufficiently carried out. However, laboratory studies of the parameter values and physical processes determining the behaviour of radon in soil and entry into dwellings has meanwhile been begun (de Meijer, 1991).

A model developed by TNO - in which the exposure to gamma radiation is also included - calculates the steady-state situation; the KVI of the University of Groningen has developed a dynamic model for radon. The KVI model has been used for verification of the measured radon concentrations in the test house in Roden. The TNO model has been used for developing regulatory standards for the radioactivity in building materials and for evaluating the effects of control measures. Since relatively little research has been conducted in the Netherlands into the effect of measures, the results are only exploratory. A combination of a high ventilation rate in the crawl space and a well-sealed floor slab seems to be the most cost-effective.

Possible future developments in the individual and collective doses due to ²²²Rn in indoor air have been discussed in this chapter. Additional measures for future housing can possibly lead to a dose reduction of 0.5 mSv a⁻¹ per occupant. Larger reductions can be achieved in existing housing. The number of houses involved depends on a remedial

action level still to be formulated for the annual average ²²²Rn concentration. For an action level of, say, 50 Bq m⁻³ and using the results of the KVI survey, the number of houses requiring remediation would be 480,000. The expected average reduction in the individual annual effective dose equivalent is of the order of 1-1.5 mSv, depending on the remedial action level adopted. The effect of control measures in dwellings with much higher-than-average ²²²Rn concentrations on the collective dose due to ²²²Rn in the Netherlands is marginal, the reduction being about 0.5% at 100 Bq m⁻³ and higher levels.

A few gaps in our knowledge of the indoor radon problem to which attention should be paid in the future are:

- testing of control measures under Dutch conditions;
- research into the interrelationship of soil parameters determining the exhalation rate;
- model validation through comparison with field measurements;
- formulation of a monitoring strategy for identification of dwellings with unusually high ²²²Rn concentrations.

9 ECONOMIC CONSEQUENCES OF RADON CONTROL MEASURES FOR THE CONSTRUCTION INDUSTRY

9.1 ECONOMIC ASPECTS OF RADON CONTROL MEASURES IN DWELLINGS

The health risk from radon (see Chapter 7) can be controlled by limiting the increased radon exposure associated with living (residing) in buildings (see Chapters 5 and 8). This can be achieved by imposing limits on the radon source strengths in indoor environments, particularly from building materials, and by lowering the infiltration rate of radon from the ground into the building interior. A few premises are given in the policy paper "Radiation protection and risk management" (VROM, 1991).

Radiation policy overlaps with housing policy, as laid down in building decrees, and shows interaction with policies in other areas, such as the reduction and reuse of residues from industrial processes (memorandum "Prevention, reuse and recycling of wastes"; VROM, 1988) and a nature conservation memorandum (RWS, 1987). The latter memorandum provides for the discontinuation of gravel extraction in the Netherlands (within 10 years), and suggests limiting the extraction of other minerals, such as clay, sand and marl. In particular, this will encourage the use of other materials, such as crushed rubble, as a substitute for gravel, and the importation of natural products such as granite, basalt and quartz.

To date, only a position on restrictive measures for the use of building materials in the construction of new dwellings has been laid down. It consists of continuing a "standstill" principle with regard to radiation levels indoors (gamma radiation and radon) from building materials currently in common use until a reduction objective has been formulated for the radiation dose both from radon infiltration and from building materials (VROM, 1991). The adoption of the standstill principle, meaning that the occupants of new (replacement) houses may not receive a higher annual dose indoors from building materials than would be the case in representative postwar houses, has led to the formulation of radioactivity limits (see 8.1.4). The limits have been calculated for the main types of material used in the Netherlands (concrete, sand-lime brick, aerated concrete and gypsum products) (van Heijningen and Ackers, 1990). A limitation per material instead of a general limit encompassing all building materials was chosen, so as to prevent a sharp increase within the general limit being permissible (no "filling") for materials making a relatively small radiation contribution. At the same time, this also strengthens the assumption that the average radiation contribution from these materials remains the same.

No position has yet been taken on the application of restrictive measures for radon in existing houses. The same is true of buildings other than houses (schools, offices, factories).

Little is as yet known about the economic consequences of radon control measures. To gain some insight into this matter, the costs and benefits of these measures will be analyzed and, if possible, quantified, to get an indication of the degree of relevance.

Radon control measures should desirably lead to reduction of the calculated number of deaths due to radiation induced tumours.

The proposed limitation of the radioactivity in building materials in accordance with the standstill principle does not give a reduction. Dose reduction in the future could possibly be achieved in three ways (however, comprehensive research has not yet been carried out):

- reducing the average radiation dose from building materials used in the construction of new houses;
- reducing, also in new construction, the radon infiltration rate into the house from the underlying soil;
- reducing infiltration rates in existing houses having much higher-than-average ²²²Rn concentrations.

For the Netherlands, the average radiation dose from building materials can be estimated to be approximately 0.7 mSv a⁻¹, about 1/3 of which from gamma radiation (see Chapter 8). A dose reduction of 15% through selection of building materials having low radioactivity concentrations for new housing, i.e. by about 0.1 mSv a⁻¹, would require a considerable effort and, among other things, entail a shift from, for example, concrete and brick to aerated concrete and gypsum blocks. The economic consequences of this cannot yet be assessed.

The radon control measures relating to building practices (lower infiltration rates) for new houses give an average individual reduction of 0.4 mSv a⁻¹ for the occupants (see 8.4). The control of radon infiltration into existing houses with a ²²²Rn concentration of, for example, above 60 Bq m⁻³ can yield an individual dose reduction of about 1.0 mSv a⁻¹ (see 8.4). The number of houses involved in the Netherlands is about 300,000.

Using the most likely mortality risk from indoor ²²²Rn exposure in the Netherlands, 0.035 Sv⁻¹ (see 7.1.1) - for continuous exposure throughout a lifetime - and an average of 2.6 occupants per house, the decrease in the number of deaths can be calculated. Supposing that 500,000 new houses can be built in the next 10 years, and that a reduction of 0.1 mSv a⁻¹ can be realized through selection of building materials, this would eventually result in a reduction of five deaths per year. Should an average reduction of 0.4 mSv a⁻¹ be realized in these new houses by lowering radon infiltration rates, this would yield an additional annual reduction of 20 deaths due to indoor ²²²Rn. If remediation could be effected in all existing houses with elevated ²²²Rn levels (above 60 Bq m⁻³ in the example chosen), this would result in a decrease in the calculated mortality due to ²²²Rn of 35 persons per year. For comparison, the mortality from ²²²Rn in the Netherlands was estimated in Chapter 7 to be 700 per year.

The application of remedial measures in existing houses with a ²²²Rn concentration exceeding a chosen value (including their identification, see 8.4) implies measurement and verification of ²²²Rn concentrations using appropriate measurement techniques. Personnel must be trained for this purpose and new jobs created. The contruction work involved will also create some employment.

As regards the setting of product standards for building materials for radiation protection purposes, the standstill approach distinguishes the following areas:

- manufacture and process control;
- restriction of the use of industrial process residues;
- extraction of minerals (sand, gravel and cement);
- import and export of building materials.

In the manufacture of building materials, a tightening of the product limits will be necessary in certain cases, and possibly also a radioactivity related selection of raw materials. A guarantee concerning the radioactivity concentration, in the form of a certificate, may possibly be required upon delivery of the raw materials. This is a new field for many manufacturers. It can be expected that this will raise the cost of the products. However, no pertinent data are yet available.

It is conceivable that the system of regulatory standards will stimulate the search for alternative building materials. This too can cause a shift within the building materials market.

One consequence of the above-mentioned restrictions in the manufacture of building materials can be that there are fewer possibilities for the reuse of residues from industrial processes. This can result in increased costs for storing these residues, with consequences for landscape management, and limitation of a logistical problem.

As a result of a possible limitation on the use of industrial by-products in the manufacture of building materials, the price of the products can gradually rise because natural raw materials will become scarcer. Curtailment of the extraction of natural products - desirable for reasons of scarcity and nature conservation - can also be hampered. It is almost impossible to express the effect of these aspects entirely in economic terms.

Limits on the radioactivity in building materials and raw materials approved in the Netherlands will affect across-the-border trading in these materials. The magnitude of the effect depends on the degree of harmonization between the Dutch regulations and regulations still to be formulated in other EC countries. It is still unclear whether other EC countries will issue their own regulations.

In 1990, the European Commission made only recommendations for reducing ²²²Rn concentrations in buildings. It suggested that remedial action be considered in existing buildings with concentrations higher than 400 Bq m⁻³. For future buildings, the ²²²Rn concentrations were not to exceed 200 Bq m⁻³ (EC, 1990). The contribution of building materials to these levels is so small (less than 10%) that standards for them are not being considered. In 1989 the Council of Europe formulated proposals for the guarantee of free trade between Member States, to effect in 1993. Free trade is also to apply to building materials.

Measures concerning the building structure or building practices for new dwellings can improve the quality of construction work and lead to new construction techniques. An incidental beneficial effect is also possible, such as the mitigation of dampness problems.

In general, the imposition of radon control measures will raise construction costs. In addition, there will be (small) extra costs for maintenance and, in some cases, increased heating costs (see 9.2). Assuming that the occupant, who experiences the benefit, bears the expenses, then this will affect the pattern of spending within a household. In public

and subsidized housing - approximately 50% of the newly built single-family houses - the government defrays a (large) part of the expenses. The cost of measures in existing houses could be borne by the occupants themselves, but could also be partly subsidized, analogous to the grant scheme for energy-conservation measures.

9.2 COSTS OF RADON CONTROL MEASURES

An investigation of the costs of reducing the present radiation level through selective use of building materials in the construction of new houses has not yet been made. An exploratory study of the cost effectiveness of techniques for control of indoor radon levels considered measures aimed at the crawl space and floor slab of single-family houses to be the most suitable.

In a model study of the costs of these measures, a distinction was made between future and existing housing. Besides the costs for performing radon tests to identify houses in which remedial action might be considered, the costs include:

- the purchase and installation costs of control techniques;
- maintenance costs
- energy costs (extra heating).

Initial investments are recalculated as annual costs, on the basis of annuities with an interest rate of 8% and amortization over 25 years. Depending on the original radon concentration in the dwelling, an estimate is made of the cost effectiveness of control methods, expressed in mSv kf^{-1} (mSv per 1000 Dutch guilders).

The measures considered and their costs are summarized in Table 9.1. It can be stated that control of indoor radon raises the price of new construction by Dfl.500.- to Dfl.2000.- per one-family house, that is, 0.5 to 2% of the building costs. The costs for these provisions, expressed in annual costs, are about Dfl.70.- to Dfl.170.-, including incidental expenses (see 9.1). For measures in existing housing, the initial investment per house is higher. The annual costs are also higher, being about Dfl.120.- to Dfl.400.-.

9.3 SUMMARY AND CONCLUSIONS

The premise that the occupants of future houses may not receive a higher annual dose from natural sources indoors than would be the case in similar houses of recent date (standstill) has led to the formulation of limits for the radioactivity in building materials (see 8.1.4). By definition, all building materials in current use meet the standards. Therefore, adoption of this premise has no economic consequences at present for the manufacture, sale and use of these materials. This may change in the future if, for example, because of scarcity the natural raw materials become more expensive.

Table 9.1. Cost effectiveness of measures for reducing radon concentrations in Dutch single-family houses with a crawl space

measure	Initial	Annual	Costs		Cost	
	costs Dfl.	costs maintenance	e energy* Total Dfl. Dfl.	Total Dfl.		
NEW HOUSE 1 floor slab, seal joints 2 natural ventilation crawl sp. 3 concrete on soil in crawl sp.	500 500	20 10 20	0 30 0	71 84 171	0.7 - 20 0.6 - 24 0.4 - 10	
<pre>4 plastic sheeting on soil in crawl sp. 5 PUR against floor slab 6 combination of 1) and 2)</pre>	1400 1750 1000	20 20 25	0 -38 30	163 146 149	0.4 - 10 0.7 - 12 1.4 - 15	
EXISTING HOUSE 1 floor slab, seal joints 2 natural ventilation crawl sp. 3 concrete on soil in crawl sp. 4 plastic sheeting on soil		20 10 20	0 30 0	122 218 393	0.4 - 12 0.2 - 9 0.2 - 4	
in crawl sp. 5 PUR against floor slab 6 combination of 1) and 2)	2500 2100 2500	20 20 25	0 -38 30	275 179 289	0.2 - 6 0.6 - 9 0.7 - 8	

^{*} gas tariff, Dfl.0.50 per m³

Restrictions may be placed on the approval of new building materials incorporating residues from industrial processes or certain imported raw materials. These restrictions are reflected in increased manufacturing costs because of the installation of test and control systems, and application of stricter process control. The magnitude of this cost item cannot yet be predicted in broad terms, but its effect on the product price will probably be small.

However, if the test results for a particular type of raw material are such that only a portion of the available material may be used in the manufacturing process, the cost of the product will increase significantly.

Structural methods of reducing radon concentrations in future houses will probably focus primarily on preventing influx of air from the crawl space into the building interior and on increasing the natural ventilation rate in the crawl space. The resulting average reduction in individual dose will be about 0.4 mSv per year for the occupants. The costs of constructing the house will rise by Dfl.500.- to Dfl.2000.-. By taking measures in existing houses with much higher-than-average ²²²Rn concentrations, an individual dose reduction of about 1.5 mSv per year could be achieved, but at higher costs per house. The increase in the construction costs will probably not greatly affect the building activities and employment in this sector; reliable data are however not available.

^{**} the first figure relates to houses with an annual radon dose of 1.2 mSv and the second figure to houses with a 3.2 mSv dose

10 EVALUATION

10.1 FROM SOURCE TO RISK - AN OVERVIEW

In this report is the "source to risk" approach has been used (see Figure 10.1). The relevant processes and parameters have been briefly summarized in Table 10.1, with reference to the corresponding (sub)sections. A number of important assumptions in the calculation are included in the table as footnotes.

10.1.1 Sources and dispersion

Two important categories of radon sources are seen in Figure 10.1: the sources of natural origin (mainly the soil), and the sources which produce emissions due to human activities and practices. The latter includes non-nuclear industries (e.g. extraction of natural gas, production of drinking water from groundwater, manufacture of building materials) and some products from these industries (e.g. domestic use of natural gas and drinking water). These industries and products fall under the risk limits proposed in the policy paper "Radiation protection and risk management" (ORS), with a maximum permissible individual mortality risk of 10^{-6} a⁻¹ per source (VROM, 1991). However, the major contributor to the radon concentration in the Netherlands is the soil.

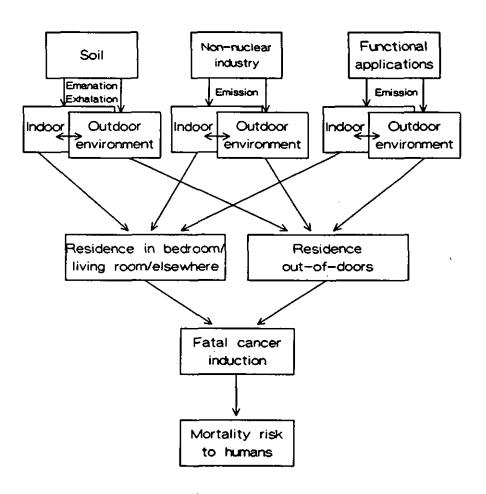


Figure 10.1. Schematic representation of the "source to risk" approach.

The parameters determining the radon exhalation rate from the soil surface are the radium and thorium concentrations, the soil porosity, the moisture content, the water table and the radon emanation coefficient. In the indoor environment, radon exhalation from building materials is, in addition to the (in)direct influence of the underlying soil, also of importance. The radon emissions from non-nuclear industries and functional applications often depend on the radium throughput in the manufacturing process.

Table 10.1. Schematic representation of the source-risk chain, with the principal processes, parameter values (and their range) and assumptions. The parameter values used in the risk calculation are given in bold face.

Position in source-risk chain	Important processes	Important parameters	Important parameters		
		Name	Value and reference		
SOURCE	NAT (a): -emanation -exhalation	• emanation	0.2 (0.01-0.8), soil (UNSCEAR, 1988)	1.2.3; 2.1	
		moisture content	~25%, soil (Kuipers, 1981)	1.1.2; 2.1	
		• porosity	0.5 (0.45-0.55); soil (Kuipers, 1981)	1.1.2; 2.1	
		diffusion	10 ⁻⁶ -10 ⁻¹⁰ m ² s ⁻² in soil	1.1.2; 2.1	
		radium content	~25 Bq kg ⁻¹ , soil (UNSCEAR, 1988)	2.1; 5.3	
		• permeability	10 ⁻¹¹ (10 ⁻⁸ -10 ⁻¹⁶) m ² (Nazaroff and Nero,1988)	1.1.2; 2.1	
		• temperature	Meteo (b)	8.1.1	
	NNI & FA (a): - emission	• radium throughput	Dependent on process	2.3; 2.4	
<u>DISPERSION</u>	INDOOR ATMOSPHERE				
	 entry into dwelling by convection and diffusion 	airtightness of floor slab	(b)	3.2.2	
		• pressure difference	(b)	8.1.1; 8.1.3	
		• ventilation	0.3 - 5 h ⁻¹	8.1.1; 8.1.3	
			Temperature difference	3.2.2	
	- release from products	• transport time	indoors/outdoors a few days? (neglected)	2.3.3	
	(natural gas, drinking water)	• transport time	a few days: (fiegrecied)	2.5.5	
	OUTDOOR ATMOSPHERE - mainly convective flow, and transport with the prevailing wind	pressure differencesprecipitationwind speed	meteorological data (b)	3.1.1; 3.1.2	

		·	, 	
EXPOSURE	INDOORS			
EXPOSURE	INDOORS - inhalation (rate)	occupancy factor living room bedroom indoors elsewhere equilibrium factor F concentration living room bedroom indoors elsewhere	0.37 (ICRP, 1987) 0.33 (ICRP, 1987) 0.18 (ICRP, 1987) 222Rn: 0.4 (de Meijer et al., 1986) 226Rn:(f) (ICRP, 1987; UNSCEAR, 1988) 227Rn: 12 Bq m ⁻³ (EEC) (de Meijer 9 Bq m ⁻³ (EEC) (c) et al.) 12 Bq m ⁻³ (EEC) (d) (1986) 226Rn: 0.5 Bq m ⁻³ (EEC)	5.5 5.5 5.5 5.4.1 5.4.1 5.4.1 5.5 5.4.1
	- external irradiation	distance from building material	(ICRP, 1987; UNSCEAR 1988)	7.1.1 5.5
	OUTDOORS - inhalation	outdoor occupancy factor	0.12 (ICRP,1987)	5.5
		equilibrium factor F concentration	²²² Rn: 0.7 (UNSCEAR,1988) ²²⁰ Rn: 0.04 (ICPR,1987) ²²² Rn: 2 Bq m ⁵ (EEC) (de Meijer et al., 1986) (e)	5.4.2 5.4.2 5.5
			²²⁰ Rn: 0.2 Bq m ⁻³ (EEC) (ICRP,1987)	5.5
EFFECT	- lung cancer induction	unattached fraction (not on earosol)	3 to 5% (ICRP,1987;Nazaroff and Nero, 1988)	6.1.1
		• quality factor for	~20 (ICRU,1986)	6.1.4
		afpha radiation dosimetric models	Various model parameters This approach yields convension coefficients for the effective dose equivalent:	6.1.4
			²²² Rn: 0.14 <u>+</u> 0.05 mSv a ⁻¹ per Bq m ⁻³ (EEC) ²²⁰ Rn: 0.6 <u>+</u> 0.2 mSv a ⁻¹ per Bq m ⁻³ (EEC)	6.1.4
<u>RISK</u>	- lung cancer mortality	epidemiology dosimetric model	²²² Rn: 3.5% per Sv ²²⁰ Rn: 5% per Sv	7.1.1 7.1.1

(a) NAT: Natural (soil, building materials); NNI: Non-nuclear industry; FA: Functional applications

Dispersion in the outdoor atmosphere depends on the meteorological conditions. For example, onshore wind "dilutes" the ²²²Rn concentration and precipitation removes radon daughters from the atmosphere by washout (see Chapter 3).

In the indoor environment, the major source of radon is entry from the underlying soil through the floor slab by convection and diffusion. In addition, convective transport (e.g., ventilation with outside air) and plateout (deposition of radon daughters on surfaces) play

⁽b) qualitative parameters

⁽c) calculated from mean F value in living rooms and mean 222Rn concentration in bedrooms

⁽d) assumption: the same as mean concentration in living rooms

⁽c) calculated from F value of the ICRP and mean 222Rn concentration in the SAWORA survey

⁽f) It can be derived theoretically that the range of F in indoor air is 0.02 - 0.1 (ICRP, 1987). Instead of an F value, the estimated EEC value of 0.5 Bq m³ was used in the risk calculations.

a role in dispersion in indoor spaces.

10.1.2 Exposures in the Netherlands

Inhalation is the major route by which radon daughters enter the body. The dose from ingestion of radon dissolved in drinking water is negligible compared with the dose from inhaled radon daughters arising from the release of radon from domestic water use (6.1.3). Following information on the average exposure from inhalation of ²²²Rn daughters and the contributions of the main sources to it, groups with elevated exposures (critical groups) and the trend in the average exposure are briefly discussed.

Average exposure

The mean outdoor atmospheric ²²²Rn concentration in the Netherlands is about 3 Bq m⁻³, and is, because of the influence of marine air, rather low compared with the concentration in continental air (average 9 Bq m⁻³).

The range in ²²²Rn concentrations observed in Dutch living rooms in the SAWORA survey was 8-140 Bq m⁻³, with a mean value of 29 Bq m⁻³ (the mean EECs are given in Table 10.1). The concentrations were consistent with a lognormal distribution. The mean concentration in the Netherlands falls within the 20-90 Bq m⁻³ range of concentrations measured in other European countries. Characteristic of the Netherlands is the absence of extreme ²²²Rn levels, as evidenced by comparison with a number of other countries. Concentrations of up to 1060 Bq m⁻³ have been measured in Dutch crawl spaces. Measurement results in buildings other than houses are not available for the Netherlands.

Model calculations indicate that building materials contribute, on average, about 8 Bq m⁻³ to the mean ²²²Rn concentration in single-family houses (8.1.3). In Table 10.2 a rough estimate has been made of the contributions of the main sources to this mean value. It shows that the soil (average about 70%, of which about 60% via infiltration and 10% via ventilation with outdoor air) and building materials (about 30%) are the dominant sources. The other sources contribute about 1% to the mean indoor ²²²Rn concentration (5.4.1; Table 2.3). The average concentrations to which the Dutch resident is exposed are about 9 Bq m⁻³ EEC (0.13 WLM a⁻¹) for ²²²Rn and 0.5 Bq m⁻³ EEC (0.088 WLM a⁻¹) for ²²⁰Rn (*).

<u>Table 10.2.</u> Contributions of the main sources to the mean indoor ²²²Rn concentration in the Netherlands

Source	Average com Bg m ⁻³	Reference	
		*	(11-4)
Soil (via infiltration)	~17	~60	(calculated)
Soil (ventilation with outside air)	3.3	11	5.4.2
Building materials	~8	~30	8.1.3
Domestic water use	0.14	0.5	5.3; Table 2.3
Natural gas	< 0.11	< 0.4	Table 2.3
Total	29	100	5.4.1

(*) There is virtually no information about ²²⁰Rn concentrations in the Netherlands. The value given for ²²⁰Rn is based on the literature (see Table 10.1).

Critical groups

The SAWORA survey found that about 0.7% of the living rooms investigated had ²²²Rn concentrations of 100 Bq m⁻³ or more. This implies that about 100,000 persons receive exposures at least three times larger than the average. Too little is still known about the critical groups among the working population. The policy document proposes is a dose limit of 2 mSv a⁻¹ for these groups (VROM, 1991). Possible groups are spa workers and persons occupationally exposed in crawl spaces, cellars, tunnels or storage facilities for uranium-bearing ores. It is not known whether a proportion of these workers receives by virtue af their profession more than 2 mSv a⁻¹.

Trend in the exposure

Based on the available data it is estimated that the average exposure to ²²²Rn around the year 2020 may be about 15% higher than the current mean value of 29 Bq m⁻³ in living rooms (see 8.2) This estimation assumed that when compared with the houses built after 1970 no important changes have occured in construction practices, building materials and ventilation rates. However, there are a number of independent developments which can influence the percentage mentioned (8.4.1).

10.1.3 Effects of and risks to humans

There is still much uncertainty about the risks to man from chronic exposure to relatively low radon concentrations. In the risk evaluation in this document, a linear dose-effect relationship without threshold has been assumed; the dose conversion coefficients for continuous exposure to 222 Rn, 220 Rn and daughters are 0.14 \pm 0.05 mSv a⁻¹ per Bq m⁻³ (EEC) (10 \pm 4 mSv WLM⁻¹) and 0.6 \pm 0.2 mSv a⁻¹ per Bq m⁻³ (EEC) (3 \pm 1 mSv WLM⁻¹), respectively. For the estimate of the lifetime risk of lung cancer mortality due to a lifetime exposure to ²²²Rn daughters (based on data from epidemiological studies of underground miners) and ²²⁰Rn daughters (based on lung dosimetry), average values of 3.5% per Sv (350 x 10⁻⁶ WLM⁻¹) and 5% per Sv (150 x 10⁻⁶ WLM⁻¹) have been used (7.1.1). The uncertainties in the overall risk calculation come to about a factor of 2. The mean annual dose has been divided according to residence area in Table 10.3. The total mean dose of about 1.7 mSv a-1 is equivalent to an average risk for a member of the population of 6x10⁻⁵ a⁻¹. This corresponds to about 700 (range: 350-1400; see 7.1.4) ²²²Rninduced and about 200 (70-600); see 7.1.4) ²²⁰Rn-induced cancer deaths per year, so that the total number of deaths due to inhalation of radon daughters in the Netherlands is approximately 900 (420-2000) annually. The uncertainty in this value comes to about a factor of 2. Table 10.4 summarizes the principal causes of the uncertainty in the risk estimate. It shows that, for ²²²Rn, this estimate is chiefly determined by the uncertainty in the risk coefficient and, to a lesser degree, by the uncertainty in the exposure estimate. The latter is important for ²²⁰Rn, however. It was furthermore assumed that a linear doseeffect relationship without threshold is applicable for the exposure of the population to radon daughters.

Table 10.3. Mean annual doses and risks from ²²²Rn and ²²⁰Rn in the Netherlands, specified by residence area

Residence area	Occupancy	222 _{Rn}		²²⁰ Rn	
	factor	concentration	dose (mSv a ⁻¹)	concentration [(Bq m ⁻³ (EEC)]	dose (mSv a ⁻¹)
At home:					
living room	0.37	12	0.62	0.5	0.11
- bedroom	0.33	9	0.42	0.5	0.10
Indoors elsewher	e 0.18	12	0.30	0.5	0.05
Outdoors	0.12	2	0.03	0.2	0.01
Total:	1.00	~9	~1.4	~0.5	~0.27
Annual risk		4.6x10 ⁻⁵		1.3x10 ^{.3}	

A firm conclusion on the synergistic relationship between radon and smoking on the basis of epidemiological studies of miner cohorts is not yet possible (6.1.7). The BEIR IV Committee found a strong synergistic interaction, but according to the BEIR V Committee the data are much too sparse to support this. In addition, there is the problem of transfer of miners' data to the general population.

There are no epidemiological studies concerning ²²⁰Rn exposures, so that conclusions are based entirely on lung dosimetry.

<u>Table 10.4.</u> Principal uncertainties in the estimate of the average risks from radon in the Netherlands

Radon isotope	Uncertainty in the exposure	Uncertainty in the concersion factor exposure → risk	Risk uncertainty	Principal causes of the risk uncertainty	
²²⁰ Rn ("thoron")	factor 2 (*)	factor 2	factor 3	uncertainty in lung dose (~30%) uncertainty in risk coefficient (dose → risk; factor 2) exposure poorly known	
²²² Rn	4-5%	factor 2	factor 2	risk coefficient for miners applied to general public	

^(*) The results of the limited Dutch observations are in good agreement with the UNSCEAR and ICRP data for countries having a moderate climate, but the sampling (N=6!) was not representative. It is assumed that the UNSCEAR and ICRP values differ by a factor of 2 at the most, from the Dutch situation.

In addition to the lung cancer risk to humans from radon exposure, there are also indications that radon can induce leukaemia and mammary carcinomas (6.1.3). The risk coefficients are still very uncertain.

10.1.4 Risks to ecosystems

Detrimental effects of radon on ecosystems, in terms of possible extinction of species, are not known (6.4).

10.2 EXCEEDING OF THE CURRENT STANDARDS AND GUIDELINES

10.2.1 Radon in outdoor air

Radon in the outdoor atmosphere arises from the decay of the naturally occurring ²³⁸U and ²³²Th in the earth's crust, and is thus one of the sources which environmental policy has categorized as "uncontrollable sources". Therefore, it is not considered practicable to set risk limits for this source. Limits have however been proposed in ORS for the added risk due to human activities; these involve discharges of radioactive materials into air and water (VROM, 1991). The maximum permissible individual risk is 10⁻⁶ a⁻¹ per source after summation of the risks from all emitted radionuclides. This implies that sources which discharge only radon into the atmosphere must, according to ORS, also meet this criterion. Examples are extraction of natural gas, some processing industries, and the storage of ores, ore tailings or phosphate fertilizers. Little is as yet known about the risks they pose in the Netherlands. Based on preliminary evaluations and information available abroad, however, it is virtually certain that radon does not significantly exceed the risk limit mentioned above.

10.2.2 Radon in indoor air

In the indoor environment, there is an extra exposure to radon above the natural outdoor level. There are as yet no hard and fast rules on quality objectives for "Building construction and occupancy", but the standstill principle is in effect used. This means that the radiation dose in future dwellings due to building materials may not be greater than the dose received in existing, representative dwellings as a result of the use of new building materials and industrial by-products. This does not rule out the possibility of average radon concentration rising, because the concentrations in new housing (or houses replacing old ones) are, on average, higher than in existing houses (see 8.2). In the future, policy will probably concentrate on reduction of the radon exposure, prevention of high radon levels in new housing and, possibly, remedial action in existing high-risk houses. Chronologically, the following steps can be distinguished: formulation of risk objective, definition of a position on infiltration rates in new buildings, and assessment of the need for a remedial action level (VROM, 1991). Environmental quality objectives do exist for ²²²Rn released during household use of natural gas and drinking water. The associated risks fall under the risk limit proposed in ORS of a maximum permissible individual risk per source of 10⁻⁶ a⁻¹ (VROM, 1991).

On an international level, there are a large number of guidelines and action levels for the concentrations of ²²²Rn in indoor air. A distinction is usually made between existing and future buildings. Action levels of 200 Bq m⁻³ (EEC) and 100 Bq m⁻³ (EEC) are often mentioned for ²²²Rn in existing and future buildings, respectively (see Table 1.4). There are no specific Dutch action levels. Although high levels were not found through, measurements in the Netherlands, it can be inferred from the SAWORA survey that there is a probability of about 0.01% that in existing homes the limit of 200 Bq m⁻³ (EEC) is exceeded (about 600 houses).

10.3 MEASUREMENT STRATEGIES

10.3.1 Measurement techniques for ²²²Rn in air

The etched-track detector in a cup used in the Netherlands is satisfactory and has a sufficiently low detection limit for a four-month exposure period (4.6). Another technique, based on adsorption of ²²²Rn onto activated carbon, is not very accurate and has a number of disadvantages (4.1.3). Integrating techniques could possibly offer prospects with regard to the cost per analysis.

10.3.2 Executed and future measurements

The surveys conducted as part of the SAWORA research programme have greatly increased the knowledge of ²²²Rn levels in the Netherlands, primarily concerned with living rooms, other rooms (usually bedrooms) and outdoor air. As part of the RENA research programme, information has been gathered about the mechanisms which determine the ²²²Rn concentration in indoor air.

If it is decided to take remedial action, then a future measurement strategy should be aimed at identifying houses with much higher than average ²²²Rn concentrations. A starting point could be a closer analysis of the SAWORA survey databases, utilizing the available information on the dwelling and the living habits of the occupants. In addition, the fundamental research currently in progress into the behaviour of ²²²Rn in Dutch soils and dwellings would be profitable. In particular, a better understanding of the role of the soil (geological factors, porosity, permeability, moisture content) and infiltration into buildings is essential. As regards the role of the local geology, advantage can possibly be taken of knowledge available in other countries in this field. Little attention has as yet been paid to radon in high-rise buildings.

Another part of the future measurement strategy could be monitoring in non-residential buildings, such as schools, day nurseries, offices and factories. Nothing is known about this as yet, and the long residence times in these buildings (see Table 5.8) can, in principle, lead to relatively high exposures. Accumulation of ²²²Rn due to the use of a ventilation system with recirculation of part of the air is not ruled out and requires investigation. Measurement strategy in the somewhat longer term can also focus on the expected increase in the ²²²Rn exposure (8.2), in order to gain insight into the actual developments and the results of any policy implemented. Surveys of the type conducted within the SAWORA framework are recommended.

10.4 COST EFFECTIVENESS OF RADON CONTROL MEASURES, SCENARIOS AND ECONOMIC CONSEQUENCES

10.4.1 Cost effectiveness of control measures

The knowledge of the cost effectiveness of control measures in the Netherlands is largely based on model calculations, which so far have been insufficient in proving their value in practice and, therefore, are only indicative (8.5). More field tests are necessary and can augment the reliability of the model calculations. With respect to the cost effectiveness of control measures, a distinction has to be made between new housing, where it is usually higher, and existing housing. For new houses, a high (natural) ventilation rate in the crawl

space combined with a well-sealed floor slab seems to be the most cost-effective. This also appears to be the case for existing dwellings. It may be noted that the effect of remedial measures on the average risk for the Dutch population from radon will be marginal in the short term; at a remedial action level for ²²²Rn of, for example, 100 Bq m⁻³, the risk reduction is about 0.5% (8.4.3). Only if effective measures are taken in new construction, leading to a general decrease in the radon concentrations, will the existing trend be broken and can there be a lower risk from radon in the long term (8.4.2).

10.4.2 Scenarios

Table 10.5 presents a number of potential scenarios which, in the long term, can lead to a reduction in the average ²²²Rn risk to members of the population (see 8.4). Should remedial action be considered in existing dwellings exceeding a specified concentration, then the problem of their identification must, of course, first be solved (8.4.3). Control measures in new construction appear to be the most promising as regards reduction in the average risk, provided that the expected effect is actually obtained. Remedial action in high-risk homes leads to a substantial reduction in the individual risk of the occupants, but its effect on the average risk from radon in the Netherlands is marginal.

10.4.3 Economic consequences of control measures

With respect to the indoor exposure to ionizing radiation from building materials, application of the standstill principle does not lead to economic consequences in the use of the current building materials. This principle can, however, lead to restrictions on new building materials incorporating, for example, industrial by-products or imported alternative raw materials. It is possible that a proportion of new industrial by-products will not be allowed as raw materials, so that problems in sales can arise and a costly storage will become necessary. The building costs of new houses with an increased ventilation rate in the crawl space and a well-sealed floor slab will rise by Dfl.1000.- to Dfl.2000.- per house.

Table 10.5. Scenarios for the reduction of the average risk from 222Rn to the population in the Netherlands

Scenario	Expected effect on the average risk
- No control measures. Situation in houses built after 1970 becomes representative for new housing (*)	15% higher around 2020
- Reduce infiltration rates and limit the radioactivity of building materials in new housing	0.6% lower annually
- Remedial action in existing houses having 222Rn levels above, for example, 50 Bq m ³ (**)	5% lower in the long term

^(*) Unlikely, because of expected independent trend (8.4.1)

10.5 CONCLUSIONS AND RECOMMENDATIONS

10.5.1 Conclusions

Since radon is a naturally occurring radioactive element, humans are exposed to it

^(**) A few examples have been worked out in 8.4.3

everywhere. As a result of the construction practices and building materials used there is an increased exposure indoors. The total annual ²²²Rn emission in the Netherlands is about 2x10¹⁶ Bq (see Table 2.3), the soil being the dominant source (95%). The major sources of ²²²Rn in indoor air are the underlying soil (average contribution about 70%) and building materials (about 30%). The other sources contribute about 1%, on average, to the mean indoor ²²²Rn concentration (see Table 10.2). The mean ²²²Rn concentration in the Netherlands falls in the range of about 20-90 Bq m⁻³ observed in other European countries. A striking feature of the distribution of the ²²²Rn concentrations in Dutch homes is the absence of extreme values. The measurement programmes did not find any houses exceeding the action levels of 200 Bq m⁻³ EEC (corresponding to 20 mSv a⁻¹) for existing buildings and 100 Bq m⁻³ (EEC) (10 mSv a⁻¹) for future buildings as recommended by the EC. In the present situation the average ²²²Rn exposure can be expected to gradually increase, because the exposures in new housing (or houses replacing old ones) are higher than in existing houses (see 8.2).

The epidemiological studies of population groups exposed to relatively low ²²²Rn levels published to date do not enable conclusions to be drawn about the health effects and numerical value of risk coefficients (6.1.6). The evaluation of the risk from ²²²Rn, lung cancer induction, is based on epidemiological studies among ²²²Rn-exposed underground miners. The risk coefficients for miners can be applied to the general population using dosimetric models. These models take into account the influence of, among other factors, breathing rate, aerosol size, and the fractions of attached and unattached ²²²Rn daughters (6.1).

There is still uncertainty regarding the risk from ²²⁰Rn, because of the absence of epidemiological data. Risk coefficients for ²²⁰Rn are based on dosimetric models. The uncertainty in the risk coefficients for ²²⁰Rn and ²²²Rn comes to about a factor of two. In addition to the risk of lung-cancer induction by radon, there are indications that radon can induce leukaemia and mammary carcinomas.

A linear dose-effect relationship without threshold has been assumed for both ²²⁰Rn and ²²²Rn, implying that every exposure, however small, leads to health risks. This assumption is also the most widely accepted internationally. The risk evaluation for the Netherlands leads to an estimated number of cancer deaths due to airborne radon of about 900 annually (uncertainty interval: 420-2000; 7.1.4). This corresponds to an average risk of 6x10⁻⁵ a⁻¹. There appear to be no risks from radon to ecosystems in terms of possible extinction of species.

If the remedial action level is set at 100 Bq m⁻³ for ²²²Rn, control measures would have to be taken in about 0.7% of the existing housing stock (about 42,000 houses) (8.4.3). It can be inferred from the SAWORA survey that if the action level is set at 200 Bq m⁻³, about 0.1% of the houses would require remediation. Of the available control techniques, an increased ventilation rate in the crawl space, combined with a well-sealed floor slab, appears to be the most cost-effective (8.3.3). However, these conclusions are as yet insufficiently underpinned by field tests and, therefore, are tentative.

The economic consequences of control measures, such as restrictions on new building materials and the use of industrial by-products, as well as criteria for the airtightness of

the floor slab, are probably small (9.3). Only for certain industrial by-products still to be approved can problems arise and can a costly storage be necessary. Significant effects on the volume of the building activities and employment are not expected.

10.5.2 Recommendations

In view of the relatively high risks to the population from exposure to radon and its daughters and the uncertainty about the effectiveness of control measures, recommendations for further research concerning the Dutch situation are made in this document. The main recommendations are listed below, with mention of the arguments or reference to the relevant section:

- field tests on the effectiveness of radon control measures in houses and other buildings with concentrations exceeding an action level still to be defined;
- development of specific Dutch criteria for identification of buildings with much higherthan-average ²²²Rn concentrations (is consistent with the ORS reduction objective in the long term, ALARA);
- further research into the economic consequences of radon control measures (these are still insufficiently known);
- examination of the ²²²Rn concentrations in schools, day nurseries, office buildings and factories (nothing is known about this; contribution to the total collective residence time is 18%);
- an exploratory monitoring programme for the exposures to ²²⁰Rn (although there are no indications that ²²⁰Rn causes problems in the Netherlands, so little is known about it that research on a limited scale is recommended);
- studies on the risks from ²²²Rn with respect to the induction of leukaemia and mammary carcinomas using laboratory animals (see 6.4);
- in the future, investigation of the ²²²Rn exposure in the Netherlands by means of a representative survey (at least 1000 measurements). In view of the slow changes in the mean ²²²Rn concentration, a frequency of once every 10-15 years will be adequate (at present, surveys are the only reliable tool for determining the trend in the exposure and for testing the results of the policy implemented).

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APPENDIX

UNITS USED

Bq becquerel (s⁻¹): the SI unit of activity, equal to one disintegration per second curie: the traditional unit of activity, equal to 3.7x10¹⁰ Bq

working level: the amount of potential alpha energy present in one cubic metre of air resulting in the emission of 2.08x10⁻⁵ J (arising historically from the potential alpha energy of 100 pCi in one litre of air; 100 pCi = 3.7 Bq). 1 WL equals 3700 Bq m⁻³ of ²²²Rn in radioactive equilibrium with its daughters (EEC²²²Rn), or 1 WL equals 270 Bq m⁻³ of ²²⁰Rn in radioactive equilibrium with its daughters (EEC²²⁰Rn)

WLM working level month: an exposure to one WL for one working month (170 hours), or 2.08×10^{-5} J m⁻³ x 170 h = 3.5×10^{-3} J m⁻³ h

QUANTITIES USED

 \mathbf{E}

A	activity concentration (Bq kg ⁻¹)
AMD	activity median diameter (m)
C	concentration (Bq m ⁻³)

d thickness of a (test) wall (m)

D_e effective bulk diffusion coefficient (m² s⁻¹)

DAC derived air concentration: the concentration (Bq m⁻³) of a given isotope in air which, at exposure for 2000 h a⁻¹ in mines, leads to the dose limit

exhalation rate (Bq m⁻² s⁻¹): rate of transport of radon gas from the surface of

a material to the atmosphere

EEC equilibrium equivalent concentration [Bq m⁻³ (EEC)]: that activity

concentration of ²²²Rn or ²²⁰Rn which, in equilibrium with its short-lived daughters, has the same potential alpha energy as the actual non-equilibrium mixture

f unattached fraction: the fraction of the radon daughter atoms which is not attached to the ambient aerosol

F equilibrium factor: the ratio of the equilibrium equivalent radon concentration to the actual radon concentration

k permeability: a measure of the ease of fluid flow through a porous material. Follows from Darcy's law

K proportionality factor: ratio of the lung dose per unit exposure in mines to that in homes

L diffusion length: the average distance traversed by a radioisotope due to diffusion in a given material within one half-life of the isotope under consideration

PAEC potential alpha energy concentration (J m⁻³): the PAEC of any mixture of

short-lived ²²²Rn or ²²⁰Rn daughters in air is the sum of the potential alpha energy of all short-lived daughters present per unit volume of air

- t time (s)
- t_{1/2} half-life (s): the time in which the activity of a radioactive substance decreases by 50%
- **P*** **Péclet number** (dimensionless): characterizes the relative importance of convective transport with respect to diffusive transport
- Q quality factor: a multiplying factor applied to absorbed dose to express the biological effectiveness of the radiation producing it
- V volume (m⁻³)
- ϵ porosity: the ratio of the pore volume to the total volume of the soil or building material
- $\epsilon_{\rm p}$ potential alpha energy: the potential alpha energy of a daughter nuclide in the decay chain of ²²²Rn or ²²⁰Rn is the total alpha energy emitted during the decay of this nuclide to ²¹⁰Pb or ²⁰⁸Pb, respectively
- η emanation coefficient (dimensionless): the fraction of the radon (²²⁰Rn or ²²²Rn) formed in the solid grains of the source material which enters the pore volume
- λ decay constant (s⁻¹): a constant which is inversely proportional to the half-life $(λ = ln(2)/t_{1/2})$
- μ viscosity (Pa s)
- ρ density (kg m⁻³)

alpha radiation: corpuscular radiation consisting of helium nuclei beta radiation: corpuscular radiation consisting of electrons

gamma radiation: electromagnetic radiation

ABBREVIATIONS USED

ALARA as low as reasonably achievable

BEIR Committee on Biological Effects of Ionizing Radiation, USA

CMB Central Medical Board, Finland

COGEMA Compagnie Générale des Matières Nucléaires, France

DGM Directorate-General for Environmental Protection, the Netherlands

EPA US Environmental Protection Agency, USA

ICRP International Commission on Radiological Protection, U.K.

ICRU International Commission on Radiation Units and Measurements, U.S.A.
ITRI Institute for Applied Radiobiology and Immunology (TNO), the

Netherlands

LET linear energy transfer

NBHW National Board of Health and Welfare, Sweden

NCRP National Council on Radiation Protection and Measurements, USA

NEA Nuclear Energy Agency, Paris

NIRP National Institute of Radiation Protection, Sweden NRPB National Radiological Protection Board, UK

ORS policy paper "Radiation protection and risk management"

PNL Pacific Northwest Laboratories, USA
RD Radiological Service TNO, the Netherlands

RENA Controllable forms of natural background radiation

RIVM National Institute of Public Health and Environmental Protection, the

Netherlands

RPA Radiation protection authorities, Nordic countries

RPB Radiation Protection Bureau, Canada

RPC Radiation Protection Commission, Germany

SAWORA Radiation Aspects of Indoor Environments and Related Radioecological

Problems

STRATEGO "Radiation relating to the built-up environment" (recently instigated

research programme)

TNO Netherlands Organization for Applied Scientific Research

UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation,

New York

VROM Ministry of Housing, Physical Planning and Environment, the Netherlands

WHO World Health Organization, Denmark