Scientific Annexes
ANNEX A

Exposures from natural sources of radiation

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Introduction

1. The major contribution to the various radiation exposures received by mankind comes from natural sources. These include external sources, such as cosmic rays and radioactive substances in the ground and in building materials, and internal sources resulting from the inhalation and ingestion of naturally occurring substances in air and diet. One characteristic of natural irradiation is that it has been experienced by the whole population of the world at a relatively constant rate over a long period of time.

2. Exposures from natural sources of radiation have been reviewed by the Committee in its 1958, 1962, 1966, 1972, 1977 and 1982 Reports. Since such exposures do not, to a large extent, vary with time, the changes in the estimates of doses in successive Reports have reflected greater knowledge in the field of natural radiation, including wider surveys of the levels of radioactive materials in the environment and in human tissues, as well as changes in the dosimetric models and quantities used.

3. The Committee has repeatedly reviewed the exposures from natural sources of radiation for a number of reasons including: (a) natural background radiation represents a substantial fraction of the total radiation exposure of most individuals and is generally the most important source of radiation; (b) the knowledge of the doses from natural background is necessary as a basis for comparison with man-made sources of exposure; (c) some exposures vary substantially according to human practices and some of the natural sources of radiation may be easily controllable; and (d) owing to the large variability of doses from natural sources of radiation, some individual doses may be high enough to warrant the introduction of remedial measures.

4. The purpose of this Annex is to update information on exposures to all sources of natural radiation. The range of activity concentrations of natural radionuclides and the range of exposures of people can be very wide. The discussion in this Annex is focused on the exposures associated with the higher portions of the observed distribution, i.e., with elevated levels of radiation or of radioactive materials. These can sometimes be identified in a comparison with the world average value of the parameter being considered, for example, a comparison of the external dose rate from cosmic radiation at a high altitude with the average at sea level; in some instances it may be more appropriate to consider the average for a country or geographical region; for example, radon concentrations in air indoors in a part of a country compared with the average for that country.

5. Both occupational exposures and exposures of members of the public are considered. Not considered is any type of exposure arising from a technological activity expressly designed to make use of the nuclear or radioactive properties of a substance, such as exposures to consumer products containing naturally occurring radionuclides or exposures due to uranium mining and milling. The latter, however, are dealt with in Annex B, "Exposures from nuclear power production".

6. The first chapter presents an overview of the natural sources of radiation and indicates the changes that have been made since the UNSCEAR 1982 Report. Chapter II presents a discussion of the doses resulting from the inhalation of radon and its decay products, which constitute the most important exposure from natural sources of radiation. In chapter III, exposures resulting from industrial activities are considered in depth.

1. NATURAL SOURCES OF RADIATION

7. Natural radiation sources are classified into two categories: (a) External sources of extraterrestrial origin, i.e., cosmic radiation, and radiation of terrestrial origin, i.e., the radioactive nuclides present in the crust of the earth, in building materials and in air; (b) Internal sources, comprising the naturally occurring radionuclides that are taken into the human body.

8. Table I summarizes the contributions of natural sources to the radiation exposure of human populations living in areas of normal radiation background. The mean annual effective dose equivalent is estimated to be 2.4 mSv. This refers to exposures of adults in the populations.
9. Some of the contributions to the total exposure to natural radiation background are quite constant in space and time and practically independent of human practices and activities. This is true for doses from ingestion of $^{40}$K, which is homeostatically controlled, and for doses from inhalation and ingestion of cosmogenic radionuclides, as such radioactive materials are to a first approximation homogeneously distributed over the surface of the globe. At the other end of the spectrum are exposures that depend strongly on human activities and practices and present a wide variability. Doses from indoor inhalation of radon and thoron decay products are typical: building design and practices, as well as the choice of building materials and of ventilation systems, influence indoor levels, thus implying a variation with time of the doses from radon as the techniques and practices evolve. Variability from one dwelling to another also stems from the wide range of radon entry rates from soil, which are to a large extent still unpredictable. In between those extreme types of exposures are several other types: (a) external doses from cosmic rays which, though affected by human practices and quite predictable, cannot be controlled except by moving to an area with a lower dose level; (b) doses from inhalation and ingestion of long-lived nuclides of the $^{238}$U and $^{232}$Th decay series, which represent a small contribution to the total dose from natural sources and which are relatively constant in space; (c) doses from external irradiation by terrestrial sources, which are also significantly altered by human activities and practices, especially through indoor exposure. Such doses, however, are, as a rule, smaller than those from inhalation of radon decay products and much less variable.

10. In comparison to the estimates given in the UNSCEAR 1982 Report, several changes have been made: (a) with respect to external exposures to cosmic radiation, the new estimates of the annual effective dose equivalents take into account the geographical distribution of the world population as a function of altitude and the shielding effect of building materials. As a result, the new estimates of the annual effective dose equivalents due to the ionizing and neutron components are higher by 20 $\mu$Sv and 30 $\mu$Sv, respectively; (b) regarding external exposure to terrestrial sources of radiation, the annual effective dose equivalents have been re-evaluated and increased by 60 $\mu$Sv, as a result of better knowledge of the indoor gamma absorbed doses in air; (c) annual effective dose equivalents from internal exposure to primordial radionuclides have been reassessed slightly downwards for the $^{222}$Rn and $^{210}$Po series, as well as for the decay products of $^{222}$Rn, whereas those for the short-lived decay products of $^{222}$Rn have been increased by about 300 $\mu$Sv on the basis of more comprehensive results of nation-wide indoor surveys. The net effect of these corrections is a 20% increase in the estimate of the overall annual effective dose equivalent from natural sources of radiation. The various contributions to the annual effective dose equivalent are discussed in more detail in this chapter. As in the previous reports of the Committee, the estimates of exposures from natural sources of radiation are essentially based on measurements in temperate latitudes, and on dietary and living habits also of the populations in these regions. It is recognized that exposures to populations in tropical latitudes may differ substantially from those in temperate latitudes, because of differences in environmental concentrations, and also in living and dietary habits. An effort has been made to estimate the exposures to populations in tropical latitudes, but the data base available is too small to enable a good assessment to be made.

11. Table 1 shows clearly the magnitude of the inhalation of $^{222}$Rn and its short-lived decay products, a topic discussed extensively in chapter II.

12. Exposures resulting from industrial activities that bring to the surface of the earth, or make available to the public, materials with enhanced concentrations of naturally occurring radionuclides do not significantly alter the picture presented in Table 1. These exposures, which may to a large extent be controlled, are dealt with in chapter III.

### A. COSMIC RAYS

13. The high-energy radiation that enters the earth’s atmosphere from outer space is known as primary cosmic rays. Upon interaction with the nuclei of atoms present in the air, neutrons, protons, pions and kaons (secondary cosmic rays) are produced, as well as a variety of reaction products (cosmogenic nuclides) such as $^3$H, $^7$Be, $^{10}$Be, $^{14}$C, $^{22}$Na and $^{24}$Na. The high-energy secondary cosmic rays thus formed react further with nuclei in the air to form more secondary particles (electrons and muons).

14. The doses from directly ionizing components of cosmic rays and from neutrons are examined separately.

#### (a) Ionizing component

15. In the lower atmosphere, the dose rates in air due to the ionizing component vary little with latitude but significantly with altitude, doubling approximately every 1,500 metres. Figure 1 shows the variation of absorbed dose rates in air, as derived from ionization chamber measurements aboard aircraft, for low and high latitudes and altitudes ranging from 0 to 12 km [H10]. Dose rates in air are about 30 nGy h$^{-1}$ at sea level for any latitude and increase to about 4 $\mu$Gy h$^{-1}$ at an altitude of 12 km and high latitudes [H10].

16. The majority of the world’s population lives at altitudes close to sea level. In the UNSCEAR 1982 Report (Annex B, paragraph 10), the absorbed dose rate in outdoor air from the ionizing component of cosmic rays was estimated to be 32 nGy h$^{-1}$; this value is taken to be numerically equal to the effective dose equivalent. Indoors, the doses are somewhat lower because of the shielding effect of building structures. Dose rates measured at the centre of various levels of
a 12-storey building showed a fairly smooth decline with depth, i.e., from the top storey down to the basement of the building (Table 2) [M9]. The marked change from the roof to the twelfth floor (65% transmission) was because of the filtering out of the relatively soft component of the cosmic-ray flux at sea level. The value of the remaining hard component decreased more slowly with depth. These measurements, however, which were made in a massive building, are not representative of the average situation. A shielding factor of about 0.85 has been reported in the case of a 0.2 m thick concrete layer [L6]. In the Netherlands, the following shielding factors for cosmic rays were determined [J17]: 0.82 for single homes with wooden ceilings; 0.76 for row houses and office buildings with wooden ceilings and floors; 0.50 for dwellings of any kind with concrete ceilings and floors; 0.42 for apartment buildings. Somewhat higher values, varying according to the floor number, were obtained in the Soviet Union [F10]: 0.81–0.96 in wooden houses; 0.72–0.92 in old stone buildings; 0.54–0.86 in modern buildings. A mean shielding factor of 0.8 is assumed in this Annex. The average indoor absorbed dose index rate at sea level is thus estimated to be about 26 nGy h⁻¹.

17. Using a value of one for the quality factor of the ionizing component of cosmic rays and an indoor occupancy factor of 0.8, the annual effective dose equivalent is estimated to be about 240 μSv at sea level. The doses received by populations living above sea level are higher: they are discussed later in this section.

(b) Neutron component

18. The variation with altitude and latitude of the neutron component is similar to that of the ionizing component (see Figure 11). At sea level, the neutron fluence rate is about 0.008 cm⁻² s⁻¹ [H10, H11]. On the basis of measurements of neutron energy spectra carried out in New York City in 1986 during a six-month experiment, Hajnal [H15] calculated an average absorbed dose rate of 0.8 nGy h⁻¹ and a dose equivalent rate of 3 nSv h⁻¹ for neutrons incident isotropically.
cally on both sides of a 30 cm thick tissue-equivalent slab. This translates into a quality factor of 3.8. In Japan, Nakamura et al. [N20] also derived from measured neutron energy spectra a dose equivalent rate of 3.3 nSv h\(^{-1}\), which is an overestimate of the effective dose equivalent rate, as it was determined using the conversion factors from flux density to maximum dose equivalent given in ICRP Publication 21 [114]. Nakamura et al. [N20] also indicated that the calculated spectra of O'Brien led to values of 2.2 nSv h\(^{-1}\) at the surface of a 30 cm body phantom and of 1.4 nSv h\(^{-1}\) on average over the phantom. These figures are consistent with the estimate of 2.4 nSv h\(^{-1}\) that was used in the UNSCEAR 1982 Report and reflect the variability associated with the choice of geometry used to calculate the effective dose equivalent. Neglecting the shielding effect of building structures, the annual effective dose equivalent for the neutron component is estimated to be about 20 μSv at sea level.

19. On the basis of radiobiological considerations, the ICRP issued in 1985 an interim recommendation according to which the quality factor for neutrons was to be increased by a factor of 2 [112]. This recommendation, however, has not been followed in most countries and has not been confirmed by the ICRP. The value of the quality factor for neutrons has not been modified in this report.

(c) Distribution of doses

20. The distribution of the effective dose equivalent from cosmic radiation over the globe was estimated by Bouville and Lowder [B38] using tabulated data on terrain heights on a 1° by 1° grid [G22], combined with population data [U7]. On the basis of information in [L14], [O3], [N20] and [H15], the variation of the annual effective dose equivalent from the ionizing component \(\hat{H}_i\), in μSv, as a function of altitude \(z\) (km) was expressed as

\[
\hat{H}_i(z) = \hat{H}_i(0) [0.205 \exp(-1.649z) + 0.795 \exp(+0.4528z)]
\]  

(1.1)

and, for the neutron component \(\hat{H}_n\), as

\[
\hat{H}_n(z) = \hat{H}_n(0) \exp(1.04z)
\]  

(1.2)

for \(z < 2\) km, and as

\[
\hat{H}_n(z) = \hat{H}_n(0) [1.98 \exp(0.698z)]
\]  

(1.3)

for \(z > 2\) km. The value used in [B38] for \(\hat{H}_i(0)\) is similar to the figure of 240 μSv a\(^{-1}\) adopted in this Annex, while the value for \(\hat{H}_n(0)\) is the same.

21. For the purpose of this Annex, the distribution of the effective dose equivalent from cosmic radiation over the globe was recalculated, using the annual sea-level effective dose equivalents of 240 μSv for the ionizing component and 20 μSv for the neutron component. Figure II shows the variation of the annual effective dose equivalent with altitude obtained from equations (1.1), (1.2) and (1.3). It is worth noting that the dose equivalent from the neutron component, which is small at sea level, increases more rapidly than the dose equivalent from the ionizing component and becomes more important at altitudes above 6 km. The distribution of the collective effective dose equivalent as a function of altitude, presented in Figure III, indicates that although about one half of the collective dose equivalent is received by people living at altitudes below 0.5 km, a contribution to the collective dose equivalent of about 10% is received by populations living at altitudes above 3 km. There are large countries with mountains and sea borders, like the Soviet Union and the United States, where the population-weighted dose from cosmic rays differs only slightly from the dose at sea level because the bulk of the population in each of

Figure III. Distribution of the collective effective dose equivalent from cosmic radiation as a function of altitude.  
(Based on [B38])
those countries lives at low altitudes [F10, O12]. In countries like Ethiopia, Islamic Republic of Iran, Kenya or Mexico, however, large cities are situated on elevated plateaux, accounting for relatively high exposures. Figure IV presents the distribution of the collective dose equivalent and the variation of the per caput dose equivalent as a function of latitude. The population of the northern hemisphere accounts for about 90% of the total collective effective dose equivalent. The per caput effective dose equivalent for the world’s population is found to be 355 μSv, the ionizing and neutron components contributing about 300 and 55 μSv, respectively.

(d) Elevated exposures

22. Elevated exposures result from prolonged presence at high altitudes. Populations living in high-altitude cities like Bogotá, Lhasa or Quito receive annual effective dose equivalents from cosmic radiation in excess of 1 mSv. Passengers and crew members aboard commercial aircraft are exposed to much higher dose rates, which vary according to the flight altitude and, to a smaller extent, the latitude and solar activity. Assuming that the average altitude of commercial subsonic flights is 8 km, the average dose equivalent rate would be about 2 μSv h⁻¹ from equations (1.1) and (1.3), the neutron component contributing about 60% of the total. Taking the annual number of hours spent flying by crew members to be 600 h, the corresponding annual effective dose equivalent is about 1 mSv. A small fraction of commercial transport is conducted with supersonic aircraft, which fly at an altitude of about 15 km. Dose equivalent rates received during supersonic transport have been reported to be about 11 μSv h⁻¹ on average for French aeroplanes [S64] and about 9 μSv h⁻¹ on average for British aeroplanes, with a maximum of around 40 μSv h⁻¹ [P14]. Annual dose equivalents received by the technical and cabin crew for the period 1979-1983 were on average about 2.5 mSv, with possible maxima of 15 mSv [P14]. For a given flight, doses received by passengers are about the same as in subsonic flights since the higher dose rates incurred during a supersonic flight are compensated for by the shorter travel time.

23. Using statistical data on air transportation, the collective dose equivalents incurred by the passenger and flight personnel of civil aviation may be estimated. The number of passenger kilometres flown in 1984, excluding Chinese airlines, was 1.3 × 10¹² [115]. Assuming an average speed of 600 km h⁻¹ at an altitude of 8 km and an effective dose equivalent rate of 2 μSv h⁻¹ at that altitude, the annual collective dose equivalent is found to be 4,300 man Sv. Published estimates [V1, W17] are 3,500 man Sv for the world population and no more than 1,600 man Sv for flights in the United States, while the collective dose equivalent to the flight crews would be up to 65 man Sv. Taking the ratio of the annual collective dose equivalent to the passengers and to the flight crews in the United States as a guide, the annual collective dose equivalent for the flight crews of the world would be about 170 man Sv.

24. While travelling in space, astronauts are subjected to primary cosmic-ray particles, radiation from solar flares and the intense radiation present in the two radiation belts. Radiation doses received by astronauts represent one of the most important constraints to long-term manned space activities [B17]. Reported dose equivalents (assumed to be effective dose equivalents) to astronauts from recent United States
manned space missions range from 0.5 to 5 mSv [B18]. Similar information is available for Soviet missions [V1]. In a majority of cases, individual effective dose equivalents did not exceed 5 mSv per flight; however, for prolonged orbital flights, i.e., those lasting longer than one month, the effective dose equivalent exceeded 10 mSv and reached 55 mSv for expedition IV on board Salut-6, which lasted 175 days [V1].

2. Internal irradiation

25. A large number of cosmogenic radionuclides are produced in the stratosphere and upper troposphere by the interaction of cosmic rays with the nuclei of atoms present in the air (e.g., nitrogen, oxygen and argon). The production and distribution of these nuclides in the environment was reviewed by the Committee in the UNSCEAR 1977 Report [U2]. Only four cosmogenic radionuclides ($^3$H, $^{14}$C, $^7$Be and $^{22}$Na) are of importance from the viewpoint of radiation doses to man. Together they deliver annual doses from internal irradiation ranging from 5 to 25 μGy in the various organs and tissues of interest. The annual effective dose equivalents are estimated to be 0.01 μSv for $^3$H, 3 μSv for $^7$Be, 12 μSv for $^{14}$C and 0.2 μSv for $^{22}$Na. Because of the relative homogeneity of the cosmic-ray flux over the earth's surface, the variability of the annual doses from the cosmogenic radionuclides is expected to be low.

B. TERRESTRIAL SOURCES OF RADIATION

26. Terrestrial sources of radiation are the very long-lived radionuclides that have existed within the earth since its formation several billion years ago and have not substantially decayed. The most important of these so-called primordial radionuclides are $^{40}$K (half-life = 1.28 $10^9$ a), $^{87}$Rb (half-life = 4.7 $10^9$ a), $^{235}$U (half-life = 4.47 $10^9$ a) and $^{232}$Th (half-life = 1.41 $10^10$ a). $^{235}$U and $^{232}$Th head series of 14 and 11 significant radionuclides, respectively, (Figures V and VI) that also contribute to the doses from terrestrial sources. Other radionuclides, such as those present in the $^{235}$U decay series, have been neglected, as they contribute little to the total dose from natural background.

27. The ubiquitous presence of the primordial radionuclides and of their decay products in the environment (air, water, soil, rocks, foodstuffs) and in humans results in external and internal radiation doses.

1. External irradiation

(a) Exposure outdoors

28. Activity mass concentrations of primordial radionuclides in rocks are usually higher in igneous rocks than in sedimentary ones, while metamorphic rocks have concentrations typical of the rocks from which they are derived. There are, however, exceptions, as

![Figure V. Uranium-238 decay series.](image-url)
certain sedimentary rocks, notably some shales and phosphate rocks, are highly active. The activity mass concentrations in soil, which are directly relevant to outdoor exposure, are thought to be largely determined by the activity mass concentrations in the source rock. The average activity mass concentrations of $^{40}$K, $^{232}$U and $^{238}$Th in soil and the corresponding absorbed dose rates in air 1 m above the ground surface are given in Table 3 [B32]; the calculations are for a soil density of 1.6 g cm$^{-3}$ and a water content of 10% and are based on the assumption that all decay products of $^{238}$U and $^{232}$Th are in radioactive equilibrium with their precursors. The average outdoor terrestrial absorbed dose rate in air from gamma-radiation 1 m above the ground surface is 44 nGy h$^{-1}$. The results of a large soil sampling programme recently carried out in the United States are in fairly good agreement with the activity concentrations given in Table 3 [M10].

29. Large-scale surveys, using different methods and types of instrumentation, have been or are being carried out in a number of countries in order to estimate average national-wide exposures to outdoor external gamma-radiation. The results for 23 countries or areas, representing about one half of the world population, are summarized in Table 4. Country-averaged outdoor absorbed dose rates in air are found to range between 24 and 85 nGy h$^{-1}$, with an arithmetic mean of 55 nGy h$^{-1}$.

30. It is to be noted that the two most populated countries in the world, China and India, have recently published the results of extensive surveys. In China, about 40,000 outdoor measurements were carried out by the Ministry of Public Health from 1981 to 1985, using sodium iodide crystals and ionization chambers in 21 provinces, five autonomous regions and the cities of Beijing, Tianjin and Shanghai. Detailed preliminary results are found in a special issue of the Chinese Journal of Radiological Medicine and Protection [C19] and a summary has been prepared by Wang Qiliang et al. [W21]. Population-weighted means of the absorbed dose rates in air are presented in Table 5: the value for the country as a whole is reported to be about 80 nGy h$^{-1}$, which is in the upper range of the levels observed in the world. These results led to a large soil-sampling programme that showed that the average concentrations of $^{40}$K, $^{232}$U and $^{238}$Th in soil in China are indeed higher than the estimates by UNSCEAR of the world averages by a factor of about 2 [W21]. Independent survey programmes of outdoor gamma-radiation have also been undertaken in China by the National Environmental Protection Agency [L18] and by the Ministry of Nuclear Industry [G29]. A comparison of the results obtained for the same areas [P22] shows discrepancies that may have arisen from the energy dependence of various types of dosimeters used in the surveys and from the estimation of the cosmic-ray dose rates which were subtracted from the readings. The average absorbed dose rates in air in the cities, provinces and autonomous regions investigated by the Ministry of Public Health [W21] were found to be lower than those measured by the National Environmental Protection Agency and the Ministry of Nuclear Industry, the relative differences ranging from 11% to 63% [P22]. Since the survey of the Ministry of Public Health is the only one completed at the time of publication of
this Annex, its results have been incorporated without modification; however, it should be kept in mind that the gamma doses for China may be revised in the near future.

31. In India, 214 locations scattered all over the country were monitored with thermoluminescent dosimeters. The national mean value of the outdoor absorbed dose rate in air due to natural radiation (terrestrial sources of radiation and cosmic radiation) is estimated to be 785 μGy a⁻¹ [N19]. Assuming a mean dose rate from cosmic rays of 300 μGy a⁻¹ yields an average absorbed dose rate in air from the terrestrial component of 55 nGy h⁻¹.

32. In the UNSCEAR 1977 Report [U2], the Committee adopted a value of 50 nGy h⁻¹ for the population-weighted average absorbed dose rate in air over the world and estimated that 95% of the world’s population residing in areas of usual natural radiation live where the outdoor absorbed dose rate in air from the primordial radionuclides lies between 30 and 70 nGy h⁻¹. As shown in Table 4, the available data base is currently much larger than it was in 1977. The population-weighted average absorbed dose rate in air for the countries in Table 4 is 63 nGy h⁻¹. This is likely to be an overestimate because of the probable bias in the Chinese measurements. When the Chinese data are not taken into account, the population-weighted average absorbed dose rate in air is reduced to 53 nGy h⁻¹. An intermediate figure of 55 nGy h⁻¹ is adopted in this Annex as the mean absorbed dose rate in air outdoor from terrestrial sources of radiation.

33. There are regions in the world where the usual range of variation of outdoor terrestrial radiation doses is substantially exceeded. The best documented of those regions are in Brazil and India. In the coastal areas of Kerala and Tamil Nadu in India, there are patches of sand containing monazite with thorium concentrations ranging between 8% and 10.5% by weight. The absorbed dose rates in air in the high radiation areas of Kerala vary between 150 and 1,000 nGy h⁻¹; in Tamil Nadu, these may reach about 6,000 nGy h⁻¹ [S69]. In Brazil, outdoor absorbed dose rates of 130-1,200, 220-4,200 and 110-1,000 nGy h⁻¹ were measured in Guarapari, Meaiche and Poços de Caldas, respectively [P24].

(b) Exposure indoors

34. Knowledge of radiation levels in buildings is important in the assessment of population exposures, as most individuals spend a large amount of time indoors. Large-scale surveys of indoor exposures to gamma external radiation have recently been conducted in several European countries as well as in China; the results are summarized in Table 6. An estimation of the world distribution of exposures may be derived from the results presented in Table 6 or, as in previous UNSCEAR Reports, from the indoor-to-outdoor ratios.

35. Estimates of average indoor absorbed dose rates in air are between 23 and 120 nGy h⁻¹, most values being in the narrow range of 60-95 nGy h⁻¹ (Table 6). The arithmetic mean is about 70 nGy h⁻¹. The population-weighted mean for all countries except China is 72 nGy h⁻¹ and becomes 110 nGy h⁻¹ if China is included.

36. In previous UNSCEAR Reports, use was made of the indoor-to-outdoor ratio, under the assumption that the relationship between the indoor and the outdoor absorbed dose rates depends essentially on the type of building materials used and their origin. If the building materials are of local origin, it may be expected that the value of the indoor-to-outdoor ratio of the absorbed dose rates in air lies between 1 and 2, because of the change in source geometry and the presence of doors and windows. Calculations taking into account the thickness and the dimensions of the walls yield ratios of 1.35 for typical brick dwellings and 1.48 for concrete buildings [K11].

37. Country-averaged indoor-to-outdoor ratios, derived from the results presented in Tables 4 and 6, range from 0.8 to 2.0, with an average of 1.3. This figure, combined with an average outdoor absorbed dose rate in air of 55 nGy h⁻¹, yields an average indoor absorbed dose rate in air of 72 nGy h⁻¹, in close agreement with some of the values obtained by the first method. In this Annex, the mean indoor absorbed dose rate in air from terrestrial sources of radiation is taken to be 70 nGy h⁻¹.

38. Elevated external dose rates indoors may arise from high concentrations of natural radionuclides in building materials. These building materials may be of natural origin (concrete based on alum shale, granite, lithoid tuff) or result from industrial processes (phosphogypsum, red mud). As discussed in the UNSCEAR 1982 Report, the resulting exposures, calculated with pessimistic assumptions, range between 100 and 2,000 nGy h⁻¹.

(c) Annual effective dose equivalents from gamma terrestrial radiation

39. The value of the quotient of effective dose equivalent rate to absorbed dose rate in air is taken, as in the UNSCEAR 1982 Report, to be 0.7 Sv per Gy for environmental exposures to gamma rays of moderate energy. This value is assumed to apply equally to males and females and to the indoor and outdoor environments. Taking the outdoor occupancy factor to be 0.2, the annual effective dose equivalent from outdoor terrestrial gamma-radiation is found to be

\[ 55 \text{ (nGy h}^{-1}) \times 0.7 \text{ (Sv G}^{-1}) \times 8,760 \text{ (h a}^{-1}) \times 0.2 = 70 \mu\text{Sv} \]

For indoor exposure, using an occupancy factor of 0.8, the annual effective dose equivalent is

\[ 70 \text{ (nGy h}^{-1}) \times 0.7 \text{ (Sv G}^{-1}) \times 8,760 \text{ (h a}^{-1}) \times 0.8 = 340 \mu\text{Sv} \]

40. The total (outdoor plus indoor) annual effective dose equivalent from terrestrial radiation, averaged over the world’s population, is 410 μSv. Using the information presented in Table 3 as a guide, the contributions of ⁴⁰K and the radionuclides of the ⁴⁰U and ²³²Th decay series to the total annual effective dose equivalent from gamma terrestrial radiation would be 150, 100 and 160 μSv, respectively.
2. Internal irradiation

41. Inhalation and ingestion of naturally occurring radionuclides give rise to internal irradiation. As in the previous reports of the Committee, the absorbed doses and effective dose equivalents are derived from measured tissue concentrations, in so far as such data are available.

(a) Potassium-40

42. Potassium is an essential element that is under close homeostatic control in the body. The average mass concentration for an adult male is about 2 g of potassium per kg of body weight [A4, K12]. The isotopic ratio of $^{40}\text{K}$ is $1.18 \times 10^{-4}$ and the average activity mass concentration of $^{40}\text{K}$ in the body is about 60 Bq kg$^{-1}$. The distribution of potassium in various tissues and organs of the body [K12] has been used to determine the concentrations of $^{40}\text{K}$ in those tissues and organs and the corresponding absorbed dose rates. The highest annual absorbed dose (270 µGy) is received in red bone marrow and the lowest in the thyroid (100 µGy). The annual effective dose equivalent is estimated to be 180 µSv. The variation of the concentration of potassium in the entire body reflects the amount of relatively potassium-free adipose tissue that is present in lean body mass at different ages: it is found to vary between about 1 and 2.5 g kg$^{-1}$.

(b) Rubidium-87

43. Very little is known about the behaviour of rubidium in the environment. Limited measurements in foodstuffs and humans seem to indicate that the human body retains rubidium more so than potassium [T12], from the mass concentrations of rubidium reported on by ICRP [13], the average activity mass concentration of $^{87}\text{Rb}$ in the body is 8.5 Bq kg$^{-1}$. The assumed distribution of rubidium in various organs and tissues of the body [13] is used to calculate the resulting absorbed dose rates from $^{87}\text{Rb}$. Bone lining cells receive the highest annual dose (14 µGy) and the thyroid the lowest (3 µGy). The annual effective dose equivalent is estimated to be about 6 µSv.

(c) Uranium-238 series

44. Uranium-238 is the head of a series of 14 principal nuclides (Figure V). This can be divided into subseries in which the activity of the precursor controls to a large degree the activities of the decay products: $^{238}\text{U} \rightarrow ^{234}\text{U}; ^{234}\text{Th}; ^{228}\text{Ra}; ^{212}\text{Rn}; ^{218}\text{Po};$ and $^{210}\text{Pb} \rightarrow ^{210}\text{Po}$. For each subseries, the intakes by inhalation and by ingestion, as well as the concentrations in bone and in soft tissues, are estimated. The results are presented in Tables 7 and 8. The conversions from the activity concentrations in bone and in soft tissues to the absorbed doses are based on the models described in ICRP Publication 30 [14]. Estimates of annual absorbed doses are shown in Table 9. Presented below is a brief discussion of the exposures with an indication of what new information has become available since the publication of the UNSCEAR 1982 Report.

45. In this Annex, uranium is assumed to consist of $^{238}\text{U}$, $^{234}\text{Th}$, $^{230}\text{Pa}$, $^{234}\text{U}$. In radioactive equilibrium with $^{234}\text{Th}$, $^{230}\text{Pa}$ and $^{238}\text{U}$, so that 1 kg of uranium contains 12 MBeq of each of the four radionuclides. The contribution of $^{238}\text{U}$ and its decay products to the total dose from natural background has been neglected.

46. In the atmosphere, the main natural source of uranium, as well as of any other precursor of one of the radon isotopes, is likely to be the resuspension of dust particles from the earth [H24, H25, K21]. Taking a dust loading of about 50 µg m$^{-3}$ in surface air of populated areas and assuming an average $^{238}\text{U}$ activity mass concentration in airborne dust as in soil of 25 Bq kg$^{-1}$ (Table 3), the activity concentration in ground-level air is estimated to be about 1.2 µBq m$^{-3}$. The corresponding annual intake by adults through inhalation is approximately 0.01 Bq (Table 7). This result is applicable to all other radionuclides of the $^{238}\text{U}$ and $^{222}\text{Th}$ series that are precursors of gaseous $^{222}\text{Rn}$ and $^{220}\text{Rn}$.

47. The annual dietary intake of $^{238}\text{U}$ has been found to be about 5 Bq in areas of normal natural activity (based on results from Japan, United Kingdom, United States). Recent measurements of uranium in the diet of New York City residents confirm the validity of this value [F12]. The contribution of drinking water to the total intake by ingestion varies over a wide range [H26]. According to the results of an extensive survey carried out in the United States, involving 90,000 measurements of domestic water supplies, average concentrations of uranium in ground water, for a given state, are only slightly higher (up to a factor of 4) than in surface water, and the ratio of the activity concentration of $^{238}\text{U}$ to that of $^{232}\text{U}$ usually ranges between 1 and 3, with an average value of 1.9 [D10, H20, H21]. It can be estimated from that survey that the population-averaged $^{238}\text{U}$ concentrations in domestic water supplies range from 0.1 Bq m$^{-3}$ in several eastern states to 50 Bq m$^{-3}$ in a number of western states, the nation-wide average in ground-water supplies being 25 Bq m$^{-3}$ [D10, H21]. Uranium-238 concentrations in European mineral water are similar to those observed in United States ground water, with median values of 12 Bq m$^{-3}$ for France and 24 Bq m$^{-3}$ for the Federal Republic of Germany. If the average daily consumption of drinking water is taken to be 0.5 litre, the average annual intake from drinking water containing 25 Bq of $^{238}\text{U}$ per m$^{3}$ is about 5 Bq, which is the same value as that found for the intake from foodstuffs. This is an example of higher intake; however, the mean intake of uranium through ingestion of water is generally likely to be small in comparison with the mean dietary intake.

48. Measured values of the activity mass concentration of $^{238}\text{U}$ in bone of adults who have lived in areas with normal dietary levels are currently more abundant [F13, F16, F18, W19]; they lie in the range of 5-150 mBq kg$^{-1}$ of $^{238}\text{U}$ in dry bone. Analyses of this wider data base indicates that a better value of the average activity mass concentration is 50 mBq kg$^{-1}$, which is lower by a factor of 3 than the previous
estimate by the Committee. The activity mass concentrations of $^{238}$U in soft tissues (Table 8) are much smaller, with the exception of the lungs, and previously measured high values in kidneys have not been confirmed [F16, W19]. The estimated annual absorbed doses from the $^{238}$U subseries are up to about 1 $\mu$Gy in bone lining cells (Table 9); the corresponding annual effective dose equivalent is 5 $\mu$Sv.

(ii) Thorium-230

49. The activity intake of $^{230}$Th through inhalation, estimated in the same manner as that of $^{238}$U, is about 0.01 Bq a$^{-1}$. The dietary intake of $^{230}$Th has been measured in the United States for the first time; it is about 2 Bq a$^{-1}$ [F12]. It can be derived from these values and ICRP models [14] that inhalation accounts for about three quarters of the total uptake to the blood of $^{230}$Th.

50. Thorium is a bone seeker having a long residence time in the skeleton and is assumed to remain on the bone surfaces [14]. The distribution of $^{230}$Th in human tissues has been investigated by Wrenn and his collaborators [13, S22, S55, W9]. A typical activity of $^{230}$Th in bone is 140 mBq, resulting in activity mass concentrations of approximately 20 mBq kg$^{-1}$ in dry cortical bone and 70 mBq kg$^{-1}$ in dry trabecular bone, assuming that the deposit of thorium is proportional to the bone area. In soft tissues, representative values of the activity mass concentrations could be 300 mBq kg$^{-1}$ in lymph nodes, 20 mBq kg$^{-1}$ in the lungs, 10 mBq kg$^{-1}$ in kidneys, 7 mBq kg$^{-1}$ in liver and 0.3 mBq kg$^{-1}$ in other soft tissues. The corresponding annual absorbed doses (Table 9) range from 7 nGy to 7 $\mu$Gy, depending on the organ or tissue considered. The annual effective dose equivalent is estimated to be about 7 $\mu$Sv.

51. The doses in bone marrow and bone lining cells, as well as the effective dose equivalent, would be significantly lower if the activity of $^{230}$Th were distributed uniformly over the mass of the skeleton instead of being concentrated on the bone surfaces. The activity mass concentration of $^{230}$Th would then be about 30 mBq kg$^{-1}$ in dry trabecular and cortical bone, while the calculated annual doses in bone marrow and bone lining cells would be 0.02 and 0.4 $\mu$Gy, respectively, instead of 0.56 and 7.4 $\mu$Gy (see Table 9). The annual effective dose equivalent would be reduced from 7 to 2 $\mu$Sv.

(iii) Radium-226

52. Food is a much more important source of radium for intake and blood uptake than is inhalation. The Committee’s previous estimate of the average annual dietary intake of $^{226}$Ra in areas of normal radiation background of 15 Bq has been recently confirmed in the USSR [D11] and the United Kingdom [S56] and is to be compared with 0.01 Bq for inhalation. The contribution of drinking water to the total intake is generally small when the drinking-water supplies are drawn from surface water; however, in ground-water supplies, which in many countries serve a large portion of the population, $^{226}$Ra concentrations vary widely and levels in excess of 200 Bq m$^{-3}$ are not uncommon (see, for example, [C18] and [H20]). Concentrations in bottled mineral water from European countries range up to 1.800 Bq m$^{-3}$ and have geometric means of 7.44, 25 and 25 Bq m$^{-3}$ in Italy, France, Austria and the Federal Republic of Germany, respectively [F17, G27, M28, R8]. In the United States, the reported geometric mean of the $^{226}$Ra concentration in public water supplies is 22 Bq m$^{-3}$. This leads to an annual intake of 4 Bq, if a water consumption rate of 0.5 litre per day is assumed.

53. When radium is taken into the body, its metabolic behaviour is similar to that of calcium, and an appreciable fraction is deposited in bone [E5]. More than 70% of the radium in the body is contained in bone [15, S57], the remaining fraction being distributed rather uniformly in soft tissues. Fisemne et al. [F4] have summarized the available data from 26 countries on measured activity mass concentrations of $^{226}$Ra in human bone. The 26 countries sampled have 1.4 10$^7$ persons and thus represent about 30% of the world population. The population-weighted distribution was found to have a median of 850 mBq per kg of calcium (corresponding to 170 mBq per kg of dry bone and to 850 mBq in the skeleton) and a geometric standard deviation of 1.6. If the fraction of $^{226}$Ra distributed in the soft tissues is taken to be 17%, as given in ICRP publication 20 [15], the average activity mass concentration in human soft tissues is found to be 2.7 mBq kg$^{-1}$.

54. The annual absorbed doses in tissues have been calculated assuming that an average retention factor of 0.33 applies to $^{222}$Rn in the skeleton (and also, conservatively, in soft tissues) and that the concentration of radium and its decay products is uniform over the total mass of mineral bone. The results, presented in Table 9, show average annual absorbed doses of less than 1 $\mu$Gy in all organs and tissues, with the exception of bone lining cells. The annual effective dose equivalent resulting from $^{226}$Ra intake in normal areas is found to be about 7 $\mu$Sv.

55. Food samples grown in the high-radiation areas of India have been collected and analysed for several radionuclides, including $^{226}$Ra. The $^{226}$Ra concentrations were found to be higher in leafy and root vegetables than in fruits and fruit vegetables. The annual intake of $^{226}$Ra by populations living in the high background areas was assessed to be about 200 Bq [L19]. A similar survey performed in the Araxa-Tapira region of Brazil showed that the annual $^{226}$Ra intakes of the most exposed people ranged between 140 and 540 Bq [P25]. The corresponding annual effective dose equivalents are between 65 and 250 $\mu$Sv.

(iv) Radon-222 and its short-lived decay products ($^{218}$Po, $^{214}$Pb, $^{214}$Bi, $^{214}$Po)

56. Because of the current scientific interest in radon exposure, a detailed treatment is provided in chapter II.

57. Average annual intakes, activity mass concentrations in lung tissues and absorbed doses are presented in Tables 7, 8 and 9, respectively. The resulting annual effective dose equivalents are estimated to be, on
average. 70 μSv for outdoor exposure and 1,000 μSv for indoor exposure. The variability around these average figures is very high.

(v) Long-lived decay products of radon-222 (²¹⁰Pb, ²¹⁰Bi, ²¹⁰Po)

58. Radon-222 evaporation from the ground constitutes the main source of ²¹⁰Pb in the atmosphere. With respect to ²¹⁰Po, the volcanic output has been estimated to be about as important as ²²²Rn exhalation [L.10]. In the mid-latitudes of the northern hemisphere, the average concentration of ²¹⁰Pb in surface air is 0.5 mBq m⁻³, while that of ²¹⁰Po is about 0.04 mBq m⁻³, as shown in Table 10; this is lower than the value adopted in the UNSCEAR 1982 Report by a factor of 2.5. Assuming that the concentrations in air are the same indoors and outdoors, the annual intakes of non-smokers through inhalation would be 4 Bq of ²¹⁰Pb and 0.3 Bq of ²¹⁰Po. Cigarette smoking leads to an increase in the intake of ²¹⁰Pb and ²¹⁰Po [M.13, P.9]. A cigarette contains about 20 mBq of ²¹⁰Pb and 15 mBq of ²¹⁰Po [P.9] and both nuclides are volatile at the burning temperature of tobacco. About 10% of the ²¹⁰Pb and 20% of the ²¹⁰Po contained in the cigarette will enter the lungs with the main smoke stream [P.9]. Therefore, for a person smoking 20 cigarettes a day, the values of the estimated annual intakes is 15 Bq for ²¹⁰Pb and 20 Bq for ²¹⁰Po. In view of the short half-life of ²¹⁰Bi (5.01 d), its activity intakes are of no importance, as ²¹⁰Bi may be assumed to be in radioactive equilibrium with ²¹⁰Pb in the body tissues; the absorbed doses from ²¹⁰Bi arise mainly from the intake of ²¹⁰Pb and not from the intake of ²¹⁰Bi itself.

59. Consumption of food is usually the most important route by which ²¹⁰Pb and ²¹⁰Po enter the human body. Concentrations of ²¹⁰Pb and ²¹⁰Po are usually low in meat and milk, intermediate in cereals and vegetables, and relatively high in aquatic organisms. Annual intakes reflect the composition of the diet of the population in question and are approximately 20 Bq in the United States [H.13, H.14], 30 Bq in the United Kingdom [S.56], about 40 Bq in the Federal Republic of Germany [G.17], the USSR [L.9], India [L.13] and Italy [C.12], and about 200 Bq in Japan [O.7, T.5]. More recent data from Japan, however, seem to indicate a much lower annual intake of 11 Bq [K.18]. Concentrations of ²¹⁰Pb in drinking water are generally low [H.20, G.27] and do not usually contribute significantly to the total intake by ingestion.

60. A well-documented case of elevated intake is that of the tens of thousands of individuals living on reindeer or caribou meat in the Arctic and sub-Arctic regions of the northern hemisphere. Their main food is the meat of these animals, which contains unusually high concentrations of ²¹⁰Po because in the winter the animals graze on lichens, which accumulate ²¹⁰Pb and ²¹⁰Po. The annual intake of ²¹⁰Pb and ²¹⁰Po by the populations living on reindeer or caribou meat are about 140 Bq for ²¹⁰Pb and about 1,400 Bq for ²¹⁰Po [H.14, P.12].

61. Lead is a bone seeker that is found incorporated in mineral bone and has a long residence time in the skeleton. In continental areas, a typical activity mass concentration of ²¹⁰Pb in dry bone is about 3 Bq kg⁻¹, yielding a skeleton content of 15 Bq. The measured ratios of ²¹⁰Po and ²¹⁰Pb activity mass concentrations in bone are centred around 0.8, leading to a ²¹⁰Po skeleton content of 12 Bq.

62. About 30% of the body content of ²¹⁰Pb is found in soft tissues, with a relatively uniform distribution throughout the body. The degree of radioactive equilibrium between ²¹⁰Po and ²¹⁰Pb depends on the organ considered; it is about 0.5 in the lungs, 0.8 in red bone marrow, and definitely greater than 1 in the liver and kidneys. Additional intake due to smoking leads to increased concentrations, particularly in the lungs.

63. The absorbed doses from the ²¹⁰Pb subseries depend mainly on the highly energetic alpha particles of ²¹⁰Po, as the contribution from the beta emissions of ²¹⁰Pb and ²¹⁰Bi amounts to about 10% of the total. The estimated annual absorbed doses of non-smokers in areas of normal dietary intake (Table 9) are about 5 μGy in soft tissues and 36 μGy in bone lining cells. The annual effective dose equivalents arising from the total intake of ²¹⁰Pb, ²¹⁰Bi and ²¹⁰Po are about 120 μSv. The corresponding figure for the populations living on reindeer or caribou meat would be about 10 times higher.

(d) Thorium-232 series

(i) Thorium-232

64. Thorium-232 is the head of a series of 11 radio-nuclides (Figure VI). The ²³²Th series has been divided into three sub-series: ²³²Th itself; ²²⁸Ra → ²²⁴Ra; and ²³⁰Rn → ²²⁶Rn. For each sub-series, the intakes and concentrations are estimated as in the case of ²³⁴U and the results are presented in Tables 7 and 8. Estimates of annual absorbed doses are shown in Table 9.

65. The activity mass concentration of ²³²Th in soil is estimated to be on average 25 Bq kg⁻¹, the same as that of ²³⁴U and its decay product ²³⁰Th (Table 3). The annual intake from inhalation is estimated to be 0.01 Bq while that from ingestion, recently measured in the United States for the first time [F.12], is about 2 Bq. Wrenn and his collaborators found the activity mass concentrations of ²³²Th in the body to be lower than those of ²³⁰Th by a factor of about 2 [S.22, W.9]. On the basis of their measurements, the body content of ²³²Th would be about 80 mBq, 60% of which is in the skeleton. The activity mass concentrations adopted in this Annex and the resulting annual absorbed doses are presented in Tables 8 and 9, respectively. The annual effective dose equivalent is calculated to be about 3 μSv.

66. The dose calculations have assumed that ²³²Th remains on bone surfaces. A volume distribution would yield an annual effective dose equivalent of about 1 μSv.

(ii) Radium-228 subseries (²²⁴Ra, ²²⁸Ac, ²²⁴Th, ²²⁴Ra)

67. Radium is much more available to plants and to animals than ²³²Th, so that the activity concentrations
of $^{228}$Ra in humans are mostly due to the dietary intake of $^{228}$Ra itself and not to the decay of $^{216}$Th in the body. Radium–228 can thus be considered to be the head of a subseries in which $^{224}$Th (half-life: 1.91 a) and $^{224}$Ra (half-life: 3.66 d), both alpha emitters, are the most important contributors to the dose.

68. The annual activity intake arising from inhalation is estimated to be 0.01 Bq, while that from ingestion of foods is considerably larger, about 15 Bq in areas of normal radiation background and about 2,000 Bq in the high background area along the Kerala coast in India. Radium–228 concentrations in drinking water are comparable to those of $^{228}$Ra but not systematically correlated when individual water supplies are considered. They are low in surface water and extremely variable in ground water [H20]. The geometric mean of the $^{228}$Ra concentrations in public ground-water supplies in the United States is reported to be 85 Bq m$^{-3}$ [H21], corresponding to an annual intake of 15 Bq for a daily drinking-water consumption of 0.5 litre.

69. The Committee estimated the average $^{228}$Ra activity mass concentrations in bone (dry weight) and in soft tissues (wet weight) of the human body to be 50 mBq kg$^{-1}$ and 4 mBq kg$^{-1}$, respectively, in areas of normal background radiation [U1, U2]. Regarding $^{228}$Th, Wrenn and Singh [W9] showed that approximately 80% of the body content (about 300 mBq) is in bone.

70. The annual absorbed doses in tissues have been calculated assuming that the $^{228}$Ra activity arising from the decay of $^{228}$Ra is retained in the body and that the concentrations of $^{228}$Ra and of its decay products are uniform over the total mass of bone. The results presented in Table 9 show annual absorbed doses greater than 1 μGy in the lungs, kidneys and bone lining cells. The corresponding annual effective dose equivalent for the subseries is found to be about 13 μSv.

(iii) Radon-220 and its decay products ($^{218}$Po, $^{212}$Pb, $^{212}$Bi, $^{212}$Po, $^{208}$Tl)

71. As is the case of $^{222}$Rn, inhalation is the major pathway through which humans are exposed to $^{222}$Rn (thoron) and its short-lived decay products. In outdoor air, the few available measurements of thoron decay products [U1, Annex D, Table 23] point to an average $^{212}$Pb/$^{222}$Rn activity concentration ratio of about 0.04. If the average $^{222}$Rn concentration is taken to be 5 Bq m$^{-3}$, the average concentration of $^{212}$Pb in outdoor air, representative of the equilibrium equivalent concentration of thoron decay products, would be 0.2 Bq m$^{-3}$. The equilibrium equivalent concentration (EEC) of thoron, or radon, is that activity concentration of thoron, or radon, in radioactive equilibrium with its short-lived decay products that has the same potential alpha energy concentration as the actual mixture of decay products. Ground-level air thoron concentrations in continental areas lie in the range of 2-10 Bq m$^{-3}$ [F11].

72. The air exchange rate indoors is always much smaller than the radioactive decay constant of thoron (half-life = 55.6 s), so that the thoron concentration in room air is relatively insensitive to the value of the ventilation rate. The thoron concentration in room air is therefore mainly determined by the exhalation rate from the soil and building materials. As the diffusion length of thoron in these materials is of the order of 1 cm, it is expected that the nature of the surface layer covering the floor and the walls has a great influence on the thoron exhalation rate; however, measurements are needed to substantiate this assumption. The magnitude of the equilibrium factor between thoron and its decay products in room air has been theoretically estimated by Portendorfer et al. [P4]. The difference between the concentrations of $^{212}$Pb and $^{212}$Bi is always found to be small but there is a factor of about 10-50, depending on the ventilation rate, between the concentrations of $^{220}$Rn (or $^{210}$Po) and $^{212}$Pb (or $^{212}$Bi), the concentrations of $^{220}$Rn being higher. The unattached fraction of $^{212}$Pb and $^{212}$Bi is very small (less than 1%) [P4].

73. The number of indoor measurements of thoron decay products is small in comparison to that of radon decay products. Table 11 summarizes the available information on the potential alpha energy ratio of thoron and radon decay products measured simultaneously. The average values are around 0.5. Using this figure and assuming a typical equilibrium equivalent concentration of radon of 20 Bq m$^{-3}$ in temperate latitudes (paragraph 140), the average equilibrium equivalent concentration of thoron (Tn) is tentatively estimated to be

\[
(20 \text{ Bq m}^{-3})_{\text{rad}} \times (55.4 \times 10^{-10} \text{ J Bq}^{-1})_{\text{rad}} \times \left(\frac{(0.5 \text{ J})_{\text{Tn}}}{(1 \text{ J})_{\text{rad}}}\right) \approx 0.7 \text{ Bq m}^{-3}
\]

in temperate latitudes.

74. For equatorial regions measurements are unavailable. Because of the different domestic conditions, the indoor concentrations of thoron decay products are expected to be lower than in temperate regions. To estimate the population-weighted world average of the indoor concentrations of thoron decay products, account must be taken of the fact that the population in tropical regions is about half that in temperate latitudes. The range of possible values for that mean concentration for the world is from 0.47 Bq m$^{-3}$ (assuming that the indoor concentration in tropical latitudes is equal to zero) to 0.7 Bq m$^{-3}$ (assuming that the indoor concentration in tropical regions is the same as that in the temperate zones). It is tentatively assumed that the population-weighted world average of the indoor concentration of thoron decay products is 0.5 Bq m$^{-3}$.

75. Regarding exposure-dose relationships, like radon and its decay products (see chapter II), thoron gas is not considered separately from thoron decay products.

76. Inhaled thoron, as a noble gas, is constantly present in the air volume of the lungs at the concentration in the inhaled air; in addition, it is partly dissolved in the lung tissues. Due to its short radioactive half-life (55.6 s), however, the equilibrium solubility in tissues other than that of the lungs will not be reached. Thus, the decay of thoron and of its
very short-lived decay product $^{210}$Po leads mainly to a
dose in the lungs; the second decay product formed,
$^{212}$Pb, is mainly transferred to blood cells, kidneys,
and bone surfaces [12].

77. Values of annual absorbed doses per unit thoron
concentrations have been derived from calculations by
Jacobi and Eifeld [J6]. They are presented in
Table 12. For concentrations of thoron gas in air in the
range of 2-20 Bq m$^{-3}$, the annual doses are up to
5 $\mu$Gy in the lungs, 4 $\mu$Gy in bone lining cells and
2 $\mu$Gy in the kidneys. The corresponding annual
effective dose equivalents are less than 20 $\mu$Sv.

78. With respect to thoron decay products, the
absorbed doses per unit inhaled activity have been
adapted from calculations of Jacobi and Eifeld [J11] to
fit the value of 0.7 Sv J$^{-1}$ used in the UNSCEAR
1982 Report [U1] for the effective dose equivalent per
unit inhaled potential alpha energy. The results,
presented in Table 12, show that the lungs, bone lining
cells and kidneys are by far the most exposed organs
and tissues. The absorbed doses per unit concentration,
also shown in Table 12, have been estimated using a mean breathing rate of 0.8 m$^3$ h$^{-1}$ indoors and
1 m$^3$ h$^{-1}$ outdoors and an average occupancy factor of
0.8 indoors and 0.2 outdoors.

79. The dosimetric coefficients of Table 12 have been
used to estimate average annual doses from inhalation
of thoron decay products. Average annual doses in the
lungs are 5 $\mu$Gy outdoors and about 40 $\mu$Gy indoors.
Annual doses in other organs and tissues are lower.
The annual average effective dose equivalent resulting
from outdoor and indoor inhalation of thoron is
estimated to be 160 $\mu$Sv.

80. Because of the few data available, the distribution
of individual exposures, as well as the reliability of
the estimated mean value, is difficult to assess at
present.

C. SUMMARY

81. A summary of the various contributions to the
annual effective dose equivalent from natural sources of
radiation is given in Table 1. In order of importance,
inhalation of short-lived decay products of radon
comes first, with the average annual effective dose
equivalent estimated to be 1,100 $\mu$Sv. A detailed
discussion on the environmental behaviour of radon
and of its dosimetry is provided in chapter II. The
second most important pathway is external irradiation,
accounting for nearly 800 $\mu$Sv, divided approximately
equally between cosmic radiation and terrestrial sources.
Less significant are the ingestion of $^{40}$K (180 $\mu$Sv),
inhalation of decay products of thoron (160 $\mu$Sv) and
internal irradiation from $^{210}$Pb-$^{210}$Po (120 $\mu$Sv). The
other natural radionuclides contribute little to the total
annual effective dose equivalent, which is estimated to
be 2,400 $\mu$Sv.

82. The variability around the mean dose from
natural sources of radiation is dominated by the radon
component, as the indoor radon concentrations span
over four orders of magnitude.

II. RADON-222 AND ITS SHORT-LIVED
DECAY PRODUCTS

83. In the UNSCEAR 1982 Report, it was estimated
that inhalation of short-lived decay products of radon
($^{222}$Rn) accounts on average for about one half of the
effective dose equivalent from all natural sources of
radiation and may sometimes lead to doses high
enough to cause concern for human health. The
awareness of the potential health problems that could
be caused by radon and other pollutants in the indoor
environment has been growing steadily in the past few
years so that a large number of scientific papers,
books, reports and meetings have been devoted to
questions related to radon exposures, especially indoors
(see, for example [A17], [C24], [H27], [N24] and
[N25]).

84. Following a brief overview of the outdoor
situation, this chapter focuses mainly on indoor
concentrations. The exposure-dose relationships for
the outdoor and indoor conditions are then established,
followed by an assessment of the annual doses.

A. OUTDOOR CONCENTRATIONS

85. Radon enters the atmosphere mainly by crossing
the soil-air interface, but there is a number of other
secondary sources, such as the ocean, ground water,
natural gas, geothermal fluids and coal combustion.
The atmospheric concentrations of radon at ground
level are governed by the source term—the exhalation
rate—and by atmospheric dilution processes, which
are both affected by meteorological conditions. The
degree of radioactive equilibrium between radon and
its decay products in the atmosphere at ground level
also depends to a large extent upon the meteorological
conditions.

86. When $^{226}$Ra decays in soil particles, the resulting
atoms of $^{222}$Rn must first escape from the soil particles
to air-filled pores and move through these pores in
order to enter the atmosphere. The escape from the
soil particles to the air-filled pores is thought to be
mainly the result of recoil of the radon atoms
following the decay of $^{226}$Ra [M1]; if they lie close to
the surface of individual grains, they may be ejected
into the pores between the grains. In comparison, the
contribution from diffusion through the solid mineral
grains is less important, as most of the radon atoms
decay before escaping. The fraction of radon formed
in the soil that escapes into the pores is known as the
emanating power, coefficient, ratio or fraction; reported
values range from about 1% to 80%.

87. In order to enter the free atmosphere, the radon
gas must diffuse through the pores of the material,
and a fraction of it will reach the surface before
decaying. The diffusion path is tortuous and, of
course, some radon atoms will be ejected into closed
pores from which they cannot escape. Movement of
radon atoms may be caused by diffusion or convec-
tion. Convective movements, induced by pressure
differences created by meteorological conditions, vary
with time and cannot be readily quantified. The diffusion process is described mathematically by the definition of an effective diffusion coefficient that includes an allowance for the convoluted path. As in the UNSCEAR 1982 Report (Annex D, paragraph 58), the area exhalation rate, defined as the activity transfer rate per unit area at the soil-air interface, is expressed as

$$R = \lambda_{\text{Ra}} F_c C_{\text{soil,Ra}} \rho_{\text{soil}} L_{\text{Ra}}$$  \hspace{1cm} (2.1)

where $R$ is the area exhalation rate in Bq m$^{-2}$ s$^{-1}$; $\lambda_{\text{Ra}}$ is the decay constant of $^{222}\text{Ra}$ (2.1 $10^{-6}$ s$^{-1}$); $F_c$ is the emanating power; $C_{\text{soil,Ra}}$ is the activity mass concentration of $^{228}\text{Ra}$ in soil (Bq kg$^{-1}$); $\rho_{\text{soil}}$ is the soil density (kg m$^{-3}$); and $L_{\text{Ra}}$ is the diffusion length of radon in soil (m), which can be expressed mathematically as the square root of $\Delta_{\text{eff}}/\lambda_{\text{Ra}} \rho_{\text{soil,ps}}$, where $\Delta_{\text{eff}}$ is the effective bulk diffusion coefficient (m$^2$ s$^{-1}$) and $\rho_{\text{soil,ps}}$ is the soil porosity.

88. Published information on experimental values of R, $F_r$, $\Delta_{\text{eff}}$ and $\rho_{\text{soil,ps}}$ is summarized in Table 13. Using the representative values given in Table 13 for $F_r$, $\Delta_{\text{eff}}$ and $\rho_{\text{soil,ps}}$ and assuming a $^{228}\text{Ra}$ activity mass concentration in soil of 25 Bq kg$^{-1}$ (Table 3) and a soil density of 1.6 $10^3$ kg m$^{-3}$; the diffusion length of radon $L_{\text{Ra}}$ is approximately 1 m and the area exhalation rate is 1.7 $10^{-2}$ Bq m$^{-2}$ s$^{-1}$, in agreement with the estimated area-weighted average per unit area for soil, based on direct measurements, of 1.6 $10^{-2}$ Bq m$^{-2}$ s$^{-1}$ [W1]. Over the ocean, the area exhalation rate is of two orders of magnitude lower than that for soil.

89. The concentrations of radon in air vary depending on the place, time, height above the ground and meteorological conditions. Because the source of radon is the soil and radon has a rather short physical half-life, the radon concentration is in general constantly decreasing with height. The geographical location is important: radon concentrations are as a rule lower over locations, such as islands and Arctic areas, which have less soil capable of emanating radon than over continental temperate regions.

90. Detailed information on the variations of radon concentration at ground level over time was obtained from a six-year record of hourly radon measurements made at Chester, New Jersey, United States [F7]. The arithmetic mean for the six years of operation was found to be 8 Bq m$^{-3}$ and the hourly data and three-hour averages of radon concentrations were log-normally distributed. A seasonal pattern of a summer maximum and a winter minimum was observed over the period 1977-1982 with little variation from year to year [F7]. The seasonal maximum in August was a factor of 3 higher than the February minimum (Figure VII). The diurnal variation (Figure VIII) shows a maximum in the night and a minimum at noon. The diurnal maximum is a factor of 2 greater than the minimum. The three-hour average radon concentrations were tested for correlation with five meteorological parameters measured at the site—temperature, dewpoint temperature, wind speed, atmospheric pressure and precipitation—but no significant correlation was found [F2].

Figure VII. Seasonal variation of outdoor radon concentration at Chester, New Jersey (1977-1982 average).

[F1]
91. The compilation obtained at Chester represents the most complete available set of data on outdoor radon concentrations at ground level. Less extensive series of measurements were reviewed in the UNSCEAR 1982 Report and by Gesell [G1]. With regard to the seasonal variations, other data from the United States showed lower values in the spring than in summer and autumn [W2, C1, G3, L2]. Data from Hungary [G9] are in agreement with those obtained at Chester. Other seasonal patterns, however, were observed at Livermore, California [L1], Bombay, India [M2], Rio de Janeiro, Brazil [L2], and Japan [I8, M3, S47]; this may be due to the fact that those locations, being islands or coastal cities, are more affected by the annual wind rose.

92. The average value of outdoor atmospheric radon concentration for normal areas of the United States was estimated by Gesell [G1] to be about 9 Bq m\(^{-3}\) after the diurnal radon cycle was taken into account. This is probably representative of the continental areas in the temperate latitudes. Values lower by a factor of 2-4 are generally observed on islands and at coastal sites [C4, F3, I8, S47, W14]. In this Annex it is tentatively estimated that the annual average of the population-weighted radon concentration in outdoor air is 5 Bq m\(^{-3}\). A lower value, 3 Bq m\(^{-3}\), was adopted in the UNSCEAR 1982 Report.

93. The equilibrium factor \(F\) between radon and its short-lived decay products is defined as the ratio of the equilibrium equivalent radon concentration to the radon activity concentration:

\[
F = \left(\frac{\chi_{eq,Rn}}{\chi_{Rn}}\right)
\]

with \(\chi_{eq,Rn} = 0.105 \chi_1 + 0.516 \chi_2 + 0.379 \chi_3\)

where \(\chi_1, \chi_2, \text{ and } \chi_3\) represent the activity concentrations of polonium-218, lead-214 and polonium-214, respectively. A few studies were devoted to the assessment of the equilibrium factor \(F\) between radon and its short-lived decay products. At Chester, New Jersey, George [G2] found an average value of 0.85 at 1 m above ground. In the Federal Republic of Germany, Jacobi obtained an equilibrium factor of 0.77 at 1-10 m above ground. Cox et al. [C1], at Cincinnati, Ohio, reported an average value of 0.87, and measurements in New Jersey and New York at 0.3-1 m above ground gave an equilibrium factor of 0.79. It seems that 0.8 may be a representative value of the average equilibrium factor \(F\) at 1 m above ground. This is higher than the value of 0.6 assumed in the UNSCEAR 1982 Report. Combining the annual average of the population-weighted radon concentration in outdoor air of 5 Bq m\(^{-3}\) with an average equilibrium factor of 0.8 yields an average equilibrium equivalent concentration of radon of 4 Bq m\(^{-3}\). Annual averages of this value vary depending on the location; a typical range, excluding extreme values, is from 1 to 10 Bq m\(^{-3}\).

**B. INDOOR CONCENTRATIONS**

94. There is no basic difference in the physical behaviour of radon and its decay products in a room and in the open air; however, the different nature of some of the sources and the magnitude of the indoor exposures warrant a separate discussion. In this section, the various sources of indoor radon are first discussed and quantified in order to estimate an average and a typical range of the rate of entry of radon into a hypothetical reference house. The principal mechanisms of transport and removal of radon and radon decay products are then considered in order to estimate average radon and radon decay product concentrations in the hypothetical reference house. In the last part of this section, the results of large-scale indoor surveys of radon or radon decay products are reviewed.

1. Sources of indoor radon

95. Radon enters buildings from different sources, such as the soil or rock under or surrounding the buildings, building materials, water supplies, natural gas and outdoor air. Characterizing the radon entry rate may require consideration of the rate at which radon is generated in source materials, of the modes of radon transport through various materials and, finally, of the manner in which radon actually enters indoor air. Several reviews of indoor radon sources have recently been prepared [N1, B25, G24, N13].

96. As in the UNSCEAR 1982 Report, reference will be made to a hypothetical reference house; its dimensions and relevant parameters are given in Table 14. In the UNSCEAR 1982 Report, the radon source in the reference house was estimated to be 74 Bq d\(^{-1}\), corresponding to a radon entry rate of about 15 Bq m\(^{-3}\) h\(^{-1}\). Figure IX presents the cumulative frequency distributions of radon entry rates determined in dwellings in several countries as the product of the simultaneously measured ventilation rate and radon concentration [N8]. It appears that the radon entry rates are approximately log-normally distributed and that there is in most countries a considerable spread between the minimum and the maximum values. The median radon entry rates presented in Figure IX range from 5 Bq m\(^{-3}\) h\(^{-1}\) in the Federal Republic of Germany...
Radon can be transported into a building from the underlying soil via diffusion or via the pressure-driven flow of air through the structural elements or through openings in the structural elements.

**Distribution**. For an uncracked slab of concrete of thickness $L$ lying on the ground, the areal exhalation rate transmitted by diffusion from the underlying soil through the slab can be expressed as $[N1, C3]$

$$R_T = \frac{R}{\cosh \left( \frac{L_c}{L_{Rn,slab}} \right)} + \frac{F_{soil,ps} L_{Rn,soil}}{F_{slab,ps} L_{Rn,slab}} \sinh \left( \frac{L_c}{L_{Rn,slab}} \right) \tag{2.3}$$

where $R$ is the area exhalation rate from uncovered soil (equation 2.1). Other formulations can be found in the literature on the subject (see, for example, [B26]). Taking the values already given in preceding paragraphs for the various parameters yields

$$R_T = 1.7 \times 10^{-2} \text{ Bq m}^{-2} \text{s}^{-1} / \cosh \left( \frac{0.2}{0.15} \right) + \frac{0.15 \times 1}{0.2 \times 0.15} \sinh \left( \frac{0.2}{0.15} \right) = 1.2 \times 10^{-3} \text{ Bq m}^{-2} \text{s}^{-1} \tag{2.4}$$

Thus, only a small percentage of the activity exhaled from the soil is transmitted through 0.2 m of uncracked concrete, and this exhaled activity represents about one half of that due to the concrete itself. The corresponding radon entry rate in the reference house can be estimated as

$$U_T = R_T N S_R / V \tag{2.5}$$

or, in numerical terms,

$$1.2 \times 10^{-3} \text{ Bq m}^{-2} \text{s}^{-1} \times 3600 \text{ s h}^{-1} \times 100 \text{ m}^2 / 250 \text{ m}^3 = 1.7 \text{ Bq m}^{-3} \text{ h}^{-1} \tag{2.5}$$

The presence of cracks in the slab may considerably increase the transmission of the diffusive flux from the soil. Using a mathematical model of a cracked slab, Landman [L3] determined that 25% of the flux from uncovered soil would penetrate the slab by diffusion if a gap of 1 cm existed for every metre of slab.

**Pressure-driven flow**. The pressure-driven flow of radon-bearing air through structural elements is believed to be an important mechanism for radon entry [N1]; it is often the predominant source of radon in dwellings with elevated concentrations (see, for example, [A10], [E7], [N8], [W13] and [W8]), especially if the house is in direct contact with the ground [S65]. Flow through an intact concrete slab is likely to be negligible in comparison to flow through cracks, holes and other penetrations. In dry soil with a density of $1.6 \times 10^3$ kg m$^{-3}$, porosity of 20%, $^{226}$Ra activity mass concentration of 25 Bq kg$^{-1}$ and emanating power of 20%, the calculated equilibrium radon concentration in soil gas is 40 kBq m$^{-3}$. If it is assumed that 0.25 m$^3$ of soil gas enters a building per hour, representing about 0.1% of the total air exchange rate, the radon entry rate from that source is 40 Bq m$^{-3}$ h$^{-1}$.

Much attention has been given to the question of radon entry through convective flow in countries
such as Sweden [S7, S8], the United States [N1], Canada [E1] and the United Kingdom [O2], where a number of dwellings have been found to have unusually high radon concentrations. The radon entry rate through convective flow $R_c$ is simply the product of the radon concentration in soil gas $C_s$ and the infiltration rate from the soil $Q_s$. The radon concentration in soil gas depends on the activity mass concentration of radium in soil, its emanating power and the porosity, permeability and moisture content of the soil. Investigations of radon concentrations in soil gas in Sweden, the results of which are presented in Table 15, show that there is usually no long-distance transport of radon in the soil and that the radon concentrations in soil gas can be explained assuming an emanating power of 10-30% [A10].

102. The infiltration rate from the soil $Q_s$ depends on the degree to which indoor air is coupled to the soil, which, in turn, depends on the design and construction of the building structure, on meteorological parameters influencing radon movements in the soil and on the living habits of the occupants, which can affect the air exchange rate in the building. Mechanisms of radon transport from soil into a house having a vented crawl space are illustrated in Figure XI [N10]. The two pressure-driven flows considered are those due to the indoor-outdoor temperature difference, including a stack effect in winter, and wind.

![Schematic diagram of mechanisms related to radon transport from soil, through a vented crawl space and into a house.](image)

103. Experimental investigations in a house with a vented crawl space showed that a higher temperature difference, including an increasing infiltration rate, corresponded to a higher indoor radon concentration, suggesting that the increased infiltration rate is more than compensated for by an increase in the radon entry rate [N10]; they also suggested that the infiltration rate from the soil into the house was, in this case, more important than the infiltration rate from the walls. A high wind speed tended to reduce the indoor radon concentration, presumably by increasing the cross-ventilation of the crawl space and the infiltration rate of the living space. For this particular house, as well as for two other houses with vented crawl spaces that were investigated in the study, it was found that perhaps 50% or more of the radon released into the crawl space from the soil beneath the house entered the living space [N10]. This figure cannot, however, be applied to all types of construction: the floor of a crawl-space house, which is usually built of wood, is likely to have greater infiltration rates than the floor and walls of a basement, which are usually built of poured concrete and concrete blocks.

104. Another extensive investigation of radon entry through the soil was conducted in a single-family house with a basement; real-time measurements of the indoor radon concentration, air exchange rate, source-related parameters and meteorological factors were performed for a period of five months [N4]. The radon entry rate was calculated from the measured radon concentration and air exchange rate. Figure XII shows the results obtained during a week of that experiment with regard to the radon entry rate and the radon concentrations in the soil adjacent to the house and in the basement sump, which collects rainwater via a drain tile system surrounding the house at a level near that of the basement floor. The good correlation between the sump activity and the radon entry rate suggests that the sump is an important pathway for radon entry into the house, although it cannot account for the total radon activity [N4]. The main conclusion derived from this study is that the radon entry rate may have two components: one may be independent of the air exchange rate, acting in a way similar to entry by diffusion; the other may be proportional to the air exchange rate, behaving like pressure-driven entry [N4]. The corresponding radon entry rates in the house were calculated to be about 2 Bq m⁻³ h⁻¹ for the diffusion process and about 60 Bq m⁻³ h⁻¹ for the pressure-driven force [N1].

105. Computer modelling of radon movement of soil gas resulting from pressure differences between the underlying soil and indoor air has been carried out in Canada [E7, S9]. In a wind-tunnel study, variations in wind speed and direction, and not the wind speed or direction itself, were found to cause large variations in the radon entry rate. The average radon entry rate was found to be mainly a function of the soil permeability and only secondarily a function of the area and resistance of the house used in the computer model to soil connections. The effect of seasonal differences on radon entry rate was approximately 4 to 1 from winter to summer. Stack-effect pressure differences due to temperature differences increased by a similar factor over this period. Because the ventilation rate of the house depends largely on the stack effect, these results suggest that summer and winter concentrations should be similar unless doors and windows are opened in summer to increase the ventilation rate. For the same geometry and weather, decreasing the soil permeability from $10^{-4}$ cm² to $10^{-6}$ cm² (a factor of 1,000) decreased the average radon entry rate into the house by a factor of about 25. This suggests that the radon entry rate is roughly proportional to the square root of soil permeability [D12]. Considerably more theoretical and experimental work is likely to be required to determine entry routes and transport mechanisms from underlying soil with some degree of confidence.
Figure XII. Variation with time in a single-family house with a basement of (a) radon entry rate; (b) the radon concentration in the basement sump; and (c) in the soil adjacent to the house.

[N4]
(b) Building materials

106. A fraction of the radon activity produced by decay of $^{226}$Ra in building materials enters buildings by diffusion. The area exhalation rate can be expressed as

$$R = \lambda_{\text{Ra}} \rho_{\text{build}} C_{\text{Ra}} F_r L_{\text{Ra}} / \text{anhn} (L_h / L_{\text{Ra}}) \quad (2.6)$$

which is an equation very similar to that related to soil (equation 2.1), the only difference being the introduction of a hyperbolic term to account for the fact that diffusion takes place in a medium of finite thickness. In equation (2.6), $R$ is the area exhalation rate ($\text{Bq m}^{-2} \text{s}^{-1}$); $F_r$ is the emanating power; $\rho_{\text{build}}$ is the density of the building material ($\text{kg m}^{-3}$); $C_{\text{Ra}}$ is the activity mass concentration of $^{226}$Ra in the building material ($\text{Bq kg}^{-1}$); $L_{\text{Ra}}$ is the diffusion length ($\text{m}$); and $L_h$ is the half-thickness of a slab of building material ($\text{m}$).

107. The mass activity exhalation rate, expressed in $\text{Bq kg}^{-1} \text{s}^{-1}$ and defined as

$$\dot{R}_m = \lambda_{\text{Ra}} C_{\text{Ra}} F_r$$

is the quantity usually determined in laboratory measurements [J1]. The area exhalation rate from a wall or a floor made of building material of half-thickness $L_h$, diffusion length $L_{\text{Ra}}$ and density $\rho_{\text{build}}$ can then be expressed as

$$R = \dot{R}_m \rho_{\text{build}} L_{\text{Ra}} / \text{anhn} (L_h / L_{\text{Ra}}) \quad (2.8)$$

108. Information on the mass exhalation rate, the diffusion length, the emanating power and the $^{226}$Ra activity mass concentration in building materials is presented in Table 16. Concrete and brick are probably the two most widely used building materials, at least in the temperate latitudes. Table 16 shows that even though the $^{226}$Ra concentrations in concrete seem to be lower than those in brick, the $^{222}$Rn mass exhalation rates appear to be higher for concrete than for brick. This is because of the high values of the emanating power for concrete, with or without fly ash, in comparison to those for brick. The results presented for gypsum (Table 16) are difficult to analyse because the few measurements available give either high or low values of the emanating power. Further measurements are necessary to clarify this point.

109. Some data on the emanating power of the concrete components are given in Table 16. Cement and fly ash have low emanating powers, probably due to their crystalline nature. Differences in the moisture content of the materials studied may also account in part for the wide disparity in the results of exhalation measurements from different countries [N1]. It is worth noting that the exhalation rate from concrete should not be derived from the emanating power of its constituents, as chemical changes occur during manufacturing.

110. The adopted reference values for the parameters of interest in the calculation of the area exhalation rate from concrete and brick are indicated in Table 16. For building material with a thickness of 0.2 m, the reference area exhalation rates would be 2 mBq m$^{-2}$ s$^{-1}$ and 0.3 mBq m$^{-2}$ s$^{-1}$ for concrete and for brick, respectively. The rate of entry of radon resulting from exhalation from building materials may be expressed as

$$U_{\text{bn}} = N / V (2R_h S_T + R_p S_p) \quad (2.9)$$

where $N$ is the number of seconds per hour; $V$ is the volume of the reference house ($250 \text{ m}^3$); $S_T$ is the surface area of its floor or ceiling (100 m$^2$); $S_p$ is the surface area of its external walls (100 m$^2$); and $R_h$ and $R_p$ are the area exhalation rates from concrete and brick, respectively.

111. The rate of entry of radon from the floor and the ceiling of the reference house would amount to about 6 Bq m$^{-3}$ h$^{-1}$, while the contribution from the external walls would be about 0.4 Bq m$^{-3}$ h$^{-1}$. The contribution from radon exhalation from the building materials to the radon concentration in the reference house is thus estimated to be 6.4 Bq m$^{-3}$ if the air exchange rate is taken to be 1 h$^{-1}$.

112. Considerably greater values of the rate of entry of radon are expected to be obtained when building materials with high $^{226}$Ra concentrations and normal emanating power are extensively used. Examples of such building materials are granite, Italian tuff and alum shale lightweight concrete. Among these materials, Swedish alum-shale lightweight concrete has the highest $^{226}$Ra concentrations (about 1,300 Bq kg$^{-1}$ on average) [S19, S20] and probably the highest $^{222}$Rn mass exhalation rate (440 mBq kg$^{-1}$ s$^{-1}$) in Table 16). Assuming a density of 2.10$^3$ kg m$^{-3}$ and a diffusion length of 7.4*10$^{-3}$ m (as measured in [J2]), a 0.2 m thick slab of 100 m$^2$ would lead to a rate of entry of radon of about 80 Bq m$^{-3}$ h$^{-1}$ in the reference house.

113. Techniques for reducing the radon entry rate due to exhalation from building materials have been investigated. In Sweden, aluminium foil has been applied to the walls of houses built with aerated concrete based on alum shale [S19]; the results showed a 50% reduction in the radon entry rate. In the United States, various radon sealants have been tested under conditions representative of normal construction conditions [E2]. The radon exhalation rate was reduced from 20% to 80%, depending on the surface coating used. Because of the trapping of radon decay products in the wall, there is a relatively small increase in the external gamma dose rate [E2, M8]. It was also noted that any cracks that later developed in a sealant such as paint may lead to leaks that negate a large portion of the sealing effectiveness of the paint [M19]. Similarly, the radon exhalation rate from any unpainted areas may be increased as they offer to radon a path of least resistance in comparison with painted areas [A11].

(c) Outdoor air

114. Air exchange between the outdoor and the indoor environments brings some outdoor radon into buildings. Air exchange arises from natural ventilation through open doors and windows, mechanical ventilation and infiltration, the uncontrolled leakage of air through cracks in the building envelope. Unless otherwise stated, air exchange in this Annex is
assumed to consist of infiltration only. Outdoor radon can play a significant role in the indoor radon entry rate if the house is poorly sealed. The radon entry rate resulting from the infiltration of outdoor air may be written as

\[ \dot{U}_{\text{out}} = \chi_{\text{out}} \dot{\nu} \]  

(2.10)

where \( \chi_{\text{out}} \) is the radon concentration in outdoor air (Bq m\(^{-3}\)); \( \dot{\nu} \) is the air exchange rate (h\(^{-1}\)); and \( \alpha \) is the fraction of the air exchange rate involving outdoor air, which in all cases is close to 1. Assuming an outdoor concentration of 5 Bq m\(^{-3}\) (see paragraph 92), an air exchange rate of 1 h\(^{-1}\) and a value of 1 for \( \alpha \), the radon entry rate would be 5 Bq h\(^{-1}\) m\(^{-3}\). It is worth noting that the radon entry rate resulting from the infiltration of outdoor air is proportional to the air exchange rate.

\( \text{(d) Water} \)

115. Radon contained in water is to some extent transferred into room air as a result of agitation or heating. Radon concentrations are as a rule much lower in surface water than in ground water. In water-saturated soil with a density of 1.6 10^3 kg m\(^{-3}\), porosity of 20% and emanating power of 20%, a 226Ra activity mass concentration of 25 Bq kg\(^{-1}\) yields at equilibrium a 222Rn concentration in ground water of 25 Bq kg\(^{-1}\) \times 1.6 10^3 kg m\(^{-3}\) \times 0.2/0.2 = 4 10^4 Bq m\(^{-3}\).

In surface water, radon concentrations are expected to be similar to those of 226Ra, that is, about 10 Bq m\(^{-3}\).

116. The release of radon from water to air depends upon the circumstances in which the water is used, as the degassed fraction increases considerably with temperature [G5]. Studies of radon transfer from tap-water to indoor air reported a use-weighted transfer efficiency of 0.5-0.6 and an average water use of 0.2-0.4 m\(^3\) per day and per person [G5, P2]. Degassing of radon from tap-water has been found to lead to elevated indoor radon concentrations in Canada [M7], Finland [C2] and the United States [G6]. Taking the air-to-water concentration ratio to be typically 10\(^{-4}\) [C6], an indoor air concentration of 400 Bq m\(^{-3}\) is obtained if the radon concentration in water is 4 MBq m\(^{-3}\). High concentrations in water are usually associated with deep wells drilled in granitic areas; the highest reported concentrations in water are 14 MBq m\(^{-3}\) in Canada [M7], 77 MBq m\(^{-3}\) in Finland [S68] and about 20 MBq m\(^{-3}\) in the United States [G6, S18]. The use of aeration can reduce the radon concentration in water by a factor of about 100 [C2].

117. Measurements of radon concentrations in water have mostly been undertaken in regions where high levels were suspected. Measurements intended to estimate the weighted average radon concentration in water for a country or a community are rare. Results of that nature are only available for Finland, Sweden and the United States. In Finland, the population-weighted average has been estimated to be 25 kBq m\(^{-3}\) for drinking water distributed by water-supply plants [A2, C2], and about 60 kBq m\(^{-3}\) when the contribution of private wells is included [K4]. The corresponding value in Sweden has been found to be 38 kBq m\(^{-3}\) [K24]. In the United States, the geometric mean radon concentrations in municipal ground-water supplies in the central part of the United States and in Texas were found to be about 4 kBq m\(^{-3}\) [P3, W3] and 5 kBq m\(^{-3}\) [W3], respectively. It was tentatively estimated in the UNSCEAR 1982 Report (Annex D, paragraph 163) that between 1% and 10% of the world's population consumes water containing radon concentrations of the order of 100 kBq m\(^{-3}\) or higher, drawn from relatively deep wells. For the remainder, who consume water from aquifers or surface sources, the weighted world average concentration is probably less than 1 kBq m\(^{-3}\).

118. The radon entry rate due to water degassing may be expressed as

\[ \dot{U}_w = \chi_w \dot{Q}_w \epsilon / V \]  

(2.11)

where \( \chi_w \) is the radon concentration in water (Bq m\(^{-3}\)); \( \dot{Q}_w \) is the amount of water used per unit time (m\(^3\) h\(^{-1}\)); \( \epsilon \) is the degassing efficiency; and \( V \) is the volume of the reference house (m\(^3\)). Assuming a radon concentration of 1,000 Bq m\(^{-3}\) in tap-water, a degassing efficiency of 0.5, a water use rate of 0.07 m\(^3\) h\(^{-1}\), corresponding to 0.4 m\(^3\) per day for each of the four persons living in the reference house, the radon entry rate is estimated to be about 0.1 Bq m\(^{-3}\) h\(^{-1}\); the corresponding mean radon concentration is 0.1 Bq m\(^{-3}\) in the reference house, with a very uneven distribution within the dwelling, the highest concentrations being expected in the rooms where radon is released (bathrooms and kitchen). In any case, average radon concentrations in tap-water do not bring a significant contribution to the total radon entry rate in buildings, except, however, when the water contains high concentrations of radon. Figure XIII shows the variation of concentrations in indoor air for a house with a radon concentration in tap-water of about 2 MBq m\(^{-3}\) [G6]; the peaks are associated with periods of high use of water in the house. The integrated radon concentrations represented by each of the peaks in Figure XIII are 200-400 Bq h m\(^{-3}\). Assuming that the characteristics of the surveyed house are the same as those of the reference house yields a water use of 0.05-0.1 m\(^3\) per peak.

\( \text{(e) Natural gas} \)

119. Natural gas is sometimes mentioned as a potential significant source of indoor radon. The radon concentration in natural gas at production wells varies from undetectable values to levels of about 50 kBq m\(^{-3}\) [G7, H2, J3, W4]. The industrial processing of natural gas involves the removal of impurities and separation of hydrocarbons. Some of these hydrocarbons are bottled under pressure for sale as liquefied petroleum gas (LPG) while others may be used for fuel. Some of the radon activity contained in the processed natural gas decays during the transit time between processing and use, or while it is stored in the bottles. When natural gas is burned in houses for cooking or space heating the radon that is released may enhance the radon level indoor if the appliances are unvented. If the combustion products are vented outside the house, this radon source is negligible.
120. The radon entry rate from the use of natural gas may be expressed as

\[ U_{ng} = \chi_{ng} \frac{Q_{ng}}{V} \tag{2.12} \]

where \( \chi_{ng} \) is the radon concentration in natural gas (Bq m\(^{-3}\)); \( Q_{ng} \) is the amount of natural gas used per unit time (m\(^3\) h\(^{-1}\)); and \( V \) is the volume of the reference house \(^1\). Radon concentrations in natural gas and LPG have mainly been measured in the United States, where the average radon concentrations in either natural gas or LPG have been found to be about 1,000 Bq m\(^{-3}\) [G8, J3]. For a consumption rate of 2 m\(^3\) d\(^{-1}\) in unvented appliances, the radon entry rate in the reference house is estimated to be about 0.3 Bq m\(^{-3}\) h\(^{-1}\). The corresponding contribution to the radon concentration is 0.3 Bq m\(^{-3}\).

(f) Summary

121. Table 17 summarizes information on the estimates of radon entry rates in the reference house. The estimated arithmetic mean value obtained is 50 Bq m\(^{-3}\) h\(^{-1}\), in agreement with arithmetic means of experimental surveys [N8]. High values can be traced to a substantial flow of soil gas from the underlying soil, the use of alum shale as a building material or elevated radon concentrations in tap-water. It should be noted, however, that all other contributors remaining the same, the relative importance of the flow of soil gas is less in apartment buildings because the ratio of volume to ground-floor surface is much larger for them than for the reference house. Its relative importance is also less if dwellings are built in such a way that the contribution of the radon entry rate from underlying soil to the total radon entry rate in living areas is very small, because either the infiltration rate from the soil is negligible or the ventilation system in the basement is separate from that of the living areas of the building.

2. Indoor behaviour of radon and radon daughters

122. Radon entering indoors accumulates as a function of the air exchange rate in the building. The activity of radon that will accumulate within a building is limited by its removal through ventilation, normally expressed as the air exchange rate (measured in air changes per hour). Radon decay product concentrations are similarly limited; in addition, they can also be subject to plate-out mechanisms, not directly connected with room ventilation rate, which will keep their concentration below that of radon.

(a) Radon

123. Assuming an instantaneous and homogeneous mixing of radon in the building, the variation of the indoor radon concentration can be described by

\[ \frac{d\chi_d(t)}{dt} = \overline{U}(t) - \chi_d(t) \left( \lambda_{Rn} + \lambda_\nu \right) \tag{2.13} \]

where \( \chi_d(t) \) is the radon activity concentration at time \( t \) in Bq m\(^{-3}\); \( \overline{U}(t) \) is the radon entry rate at time \( t \) in Bq m\(^{-3}\) h\(^{-1}\); \( \lambda_{Rn} \) is the decay constant of radon, 7.6 \( 10^{-3} \) h\(^{-1}\); and \( \lambda_\nu \) is the air exchange rate at time \( t \) in h\(^{-1}\).

124. If the radon entry rate (\( \overline{U} \)) and the ventilation rate (\( \lambda_\nu \)) are constant with time, the radon concentration at equilibrium is given by

\[ \chi_e = \frac{\overline{U}}{\lambda_{Rn} + \lambda_\nu} \tag{2.14} \]
If the contributions from outdoor air and pressure-driven flows through the soil are small, the indoor radon concentration at equilibrium is, as a first approximation, inversely proportional to the air exchange rate. This is illustrated in Figure XIV, which presents data from an experiment in which a balanced mechanical system—that is, one with equal intake and exhaust flows—was used to vary the air exchange rate \( \lambda \), without affecting the pressure-driven flow from the soil [N5].

125. Equation (2.14) can only be used as a first approximation of the mean indoor radon concentration, as it is based on assumptions that are usually not strictly valid. They are as follows: (a) the radon concentration within the building is assumed to be homogeneous. There is in fact essentially universal agreement that the radon concentrations decrease as the storey numbers of a multi-storey building increase, especially between the basement, first and second floor [G1, G9, G10, S10]. In the upper storeys of high-rise buildings, the radon concentration does not always decrease as the storey number increases. The presence of direct routes, such as lift shafts and service ducts between the basement, or even the sub-basement level, and higher storeys can distort the radon concentration gradient; (b) the air exchange rate is assumed to be constant with time. Actually, it is subject to important variations, as it is a function of human activities (opening or closing doors or windows, turning ventilation systems on or off) and of meteorological conditions (wind speed and direction, temperature, pressure). Figure XV shows the measured variation of the air exchange rate in an occupied house over a few days, together with the wind speed observed at a weather station 20 km away from the house [N6]. The air exchange rate is shown to vary from 0.5 h\(^{-1}\) to more than 4 h\(^{-1}\) in a matter of a few hours; there is in that particular case a good correlation between the wind speed and the air exchange rate, as predicted by the model of Grimsrud et al. [G25]; (c) the radon entry rate is assumed to be constant with time. In fact, as shown in Figure XII, the entry rate may experience significant changes in the course of one day owing to variation with time of the different radon sources discussed in the previous section.

126. According to equation (2.14), there is a clear relationship between the radon concentration and the air exchange rate. However, simultaneous measurements of the air exchange rate and of the instantaneous radon concentration have been performed in three surveys involving 98 houses, including both energy-efficient and conventional houses. The surveys failed to reveal such a correlation between the indoor radon concentration and the air exchange rate, although each parameter varied over a wide range [N3]. Similar results were obtained from time-averaged measurements [D2] (Figure XVI).

![Figure XIV](image-url)  
Figure XIV. Average steady-state radon concentration in a residence as a function of ventilation rate. (The dashed line indicates a constant radon source of 90 Bq m\(^{-2}\) h\(^{-1}\)).

![Figure XV](image-url)  
Figure XV. Variation of the ventilation rate in an occupied house. The wind-speed data represent observations at a weather station 20 km away from the house.

[N6]
127. Even though equation (2.14) is very simple and can be used to derive a typical value of the mean indoor radon concentration (for example, $\chi = 55 \text{ Bq m}^{-3}$ with the radon entry rates given in Table 17 and $\lambda_s = 1 \text{ h}^{-1}$), it is clear that the prediction of the actual radon concentration (and of its variation as a function of time) in a given building cannot at this stage be made with accuracy. Figure XVII shows that the variation of the indoor radon concentration over one week has no consistent pattern [G11]. Further theoretical and experimental work (as in [B26]) is required to derive the values of $\chi$, $\text{U}$ and $\lambda_s$ from the measurements of environmental and technological parameters.

128. When an atom of $^{218}\text{Po}$ is formed through the decay of radon, it is a free ion. Molecules of water vapour or trace gases coalesce almost immediately around the ion, forming a molecular cluster of 2-20 nm in diameter (Figure XVIII). The ion and the cluster are usually referred to as free, uncombined or unattached decay products. Unattached $^{218}\text{Po}$ is highly mobile and, after 10-100 s, it attaches to an aerosol particle (normal size in the range of 50-500 nm; see Figure XIX); the other options offered to unattached $^{218}\text{Po}$ are to plate out on the indoor surfaces, to be transported outdoors with the outgoing air or to decay into unattached $^{214}\text{Pb}$. Attached $^{218}\text{Po}$ is relatively immobile and its plate-out on indoor surfaces is sometimes ignored. The various processes are illustrated in Figure XIX. The attached and the unattached decay products are often distinguished by their environmental behaviour and deposition pattern in the respiratory system. In a series of air samples, the position and size of the peak related to the unattached fraction may vary for reasons not yet completely elucidated [K16].

129. Upon alpha decay of attached $^{218}\text{Po}$, the decay product $^{214}\text{Pb}$ thus created may remain on the aerosol or indoor surface or become unattached as the result of its recoil energy. Subsequently, the behaviour of $^{214}\text{Pb}$ is similar to that of $^{218}\text{Po}$. Upon the decay of attached $^{214}\text{Pb}$, the $^{214}\text{Bi}$ created typically remains attached, since the recoil energy from beta decay is not sufficiently large to promote detachment. The indoor behaviour of the radon decay products is illustrated in Figure XX, where subscripts 1, 2 and 3 relate to decay products $^{218}\text{Po}$, $^{214}\text{Pb}$ and $^{214}\text{Bi}$, respectively, and superscripts fr, a and s designate the unattached state, the attached state and the presence on indoor surfaces [B1]. Dotted lines denote the processes that have been neglected in this treatment. This model of indoor behaviour of radon decay products was first proposed by Jacobi [J4] and has subsequently been extended and modified by a number

![Figure XVI. Scatter plot of radon-222 concentrations versus air exchange rate for 58 houses in four cities. Measurements were made during a four- to five-month period between November 1981 and May 1982.](image1)

![Figure XVII. Variation with time of indoor radon concentrations.](image2)

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Figure XVIII. Histograms of size distribution of radon decay products.

Figure XIX. The basic processes influencing the activity balance of radon decay products.

Figure XX. Flow chart of radon decay product behaviour under average conditions.
of researchers, notably Porstendörfer et al. [P4, P5], Shimo et al. [S46, S48, S49] and Bruno [B1]. The plate-out rate constant of unattached radon decay products can be expressed as \( k_W = v_S S/V \), where \( v_S \) is the deposition velocity (m h\(^{-1}\)). \( S \) the total surface area in the room and \( V \) the volume of the room. The deposition velocity is about 2 m h\(^{-1}\) for unattached daughters in still air [G12, K5, P5]. The surface to volume quotient \( S/V \) is estimated to range from 3 to 6 m\(^{-1}\) in rooms of dwellings if the furniture is taken into account [P5]. The plate-out rate constant would therefore be about 10 h\(^{-1}\) in still air. In ventilated rooms, the deposition velocity increases with the air speed so that the plate-out rate constant is expected to range from 10 to 30 h\(^{-1}\) in dwellings. Values reported in the literature on the subject range from 2 h\(^{-1}\) to 200 h\(^{-1}\) [I7, I9, J4, K5, K17, P4, S11, S44, W5].

130. The deposition velocity for attached daughters is approximately two orders of magnitude less than that for unattached decay products, leading to plate-out rate constants of 0.1-0.3 h\(^{-1}\) [K5, P5]. Consequently, plate-out of attached decay products is negligible as a removal mechanism relative to decay and ventilation.

131. The attachment rate \( X \) to aerosol particles has been shown by Porstendörfer and Mercer [P6] to be a linear function of the particle concentration

\[
X = \beta Z
\]  
(2.15)

where \( \beta \) is the attachment coefficient in cm\(^3\) h\(^{-1}\) and \( Z \) is the particle concentration in cm\(^{-3}\). Reported values of the attachment coefficient range from 1.8 \( \times \) 10\(^{-4}\) cm\(^3\) h\(^{-1}\) to 7.4 \( \times \) 10\(^{-2}\) cm\(^3\) h\(^{-1}\) [P5]. For indoor air, an average attachment coefficient of 5 \( \times \) 10\(^{-3}\) cm\(^3\) h\(^{-1}\) may be assumed [P5]. The aerosol concentration indoor depends mainly on the outdoor particle concentration, the ventilation rate and the indoor sources of particles. Figure XXI presents the measured variation of aerosol concentra-

132. The probability for recoil detachment from aerosol particles upon alpha decay, also called recoil factor, has been studied from the theoretical and experimental point of view by Mercer [M4]. The recoil energy possessed by a daughter product after alpha decay is about 100 keV, corresponding to a range of about 0.1 µm in solid material. For a typical aerosol of median diameter 0.2 µm, the recoil factor is about 0.8 [M4]. Upon beta decay, the recoil energy is only a few electron volts, yielding a recoil factor of 0.01-0.02 [P4]. The detachment from walls and other surfaces has not been examined in detail in any work to date; it would be expected that 25-50% of deposited activity could re-enter the room air mainstream upon alpha decay [B1, M1].

133. As already indicated, the equilibrium factor \( F \) is defined as the ratio of the equilibrium radon activity concentration to the radon activity concentration:

\[
F = \frac{\chi_{eq,\text{Ra}}}{\chi_{\text{Ra}}}
\]  
(2.16)

with

\[
\chi_{eq,\text{Ra}} = 0.105 \chi_1 + 0.516 \chi_2 + 0.379 \chi_3
\]

where \( \chi_1 \), \( \chi_2 \) and \( \chi_3 \) represent the activity concentrations of polonium-218, lead-214 and polonium-214, respectively. The unattached fraction \( f_p \) is usually defined as the ratio of the equilibrium equivalent radon concentration in the unattached state and of the total:

\[
f_p = \frac{\chi_{eq,\text{Ra}}}{\chi_{eq,\text{Ra}} + \chi_{\text{Ra}}}
\]  
(2.17)

It is also, however, sometimes defined as the ratio of the equilibrium equivalent radon concentration that is in the unattached state and of the radon concentration:

\[
f_p = \frac{\chi_{eq,\text{Ra}}}{\chi_{eq,\text{Ra}} + \chi_{\text{Ra}}}
\]  
(2.18)

The fact that two different definitions may be used for the same quantity leads to a certain amount of ambiguity in the interpretation of the results reported in the literature when the definition used is not clearly specified. The same ambiguity exists when the unattached fraction of a given decay product of radon is reported, as the denominator of the equation can be the radon activity concentration, the total equilibrium equivalent radon activity concentration, the equilibrium equivalent radon activity concentration of the decay product under consideration. In this Annex, it is assumed in all cases that the denominator of the equation consists of the equilibrium equivalent radon concentration, as, for example, in equation (2.17).

134. Bruno [B1] used the results of the survey conducted by George et al. [G11] on long-term
average concentrations of radon and radon decay product in 21 houses in the New York area to derive the best fit for the attachment rate and the plate-out constant rate. Assuming a ventilation rate of 1 h⁻¹, an equilibrium factor of 0.5 and a relatively high unattached fraction of 0.07, Bruno obtained $\lambda_{po} = 8$ h⁻¹ and $X = 45$ h⁻¹, as shown in Figure XX, and indicated that the model would be more accurately validated with the help of simultaneous measurements of ventilation rate, radon concentrations and concentrations of attached and unattached $\text{P}^{218}\text{Po}$ [B1].

135. Using another approach, Porstendörfer [P5] analysed the sensitivity of $F$ and $f_p$ to the variations of some influencing parameters. He concluded that $F$ and $f_p$ are only slightly influenced by the outdoor concentrations of radon daughters (for $\lambda_s < 1$ h⁻¹), the value of the recoil factor, the radon exhalation rate and the plate-out rate of attached decay products; however, $F$ and $f_p$ are greatly influenced by the plate-out rate of the unattached daughters and by the aerosol concentration in the room (Figure XXII). The theoretical curves shown in Figure XXII conform well to the experimental data for $\lambda_{po} = 20-30$ h⁻¹. Since in most cases the aerosol concentrations in dwellings with low ventilation rates ($\lambda_s < 0.3$ h⁻¹) vary between $5 \times 10^3$ and $10^4$ cm⁻³ (corresponding to attachment rates of 25-50 h⁻¹ [P5]), $F$ values between 0.25 and 0.4 and $f_p$ values between 0.05 and 0.1 can be expected. This is in agreement with the experimental results of Wicke [W6] and Keller et al. [K6], who measured median $F$ values of 0.37 and 0.33, respectively, in buildings in the Federal Republic of Germany. In another study, however, it was claimed that the model tends to underestimate the equilibrium factor, as measured values of the deposition rates of unattached radon decay products in occupied buildings are found to be smaller than those obtained in laboratory experiments [S44].

136. The influence of the particle concentration on the equilibrium factor is further illustrated in Figure XXIII [S43], which shows the results of a study in which the equilibrium factor increased from 0.1 to 0.8 as the particle concentration varied from 1,000 to 25,000 cm⁻³, a range typical of the indoor environment. Because of the great influence of the aerosol concentration in buildings on the equilibrium factor and on the unattached fraction, it is recommended that more simultaneous measurements of the equilibrium factor and the aerosol concentration be conducted.

137. In summary, the basic processes influencing the concentrations of the radon decay products in indoor environments are qualitatively well known. Quantitatively, it seems that the largest uncertainty lies in the determination of the plate-out rate of unattached decay products $\lambda_{po}$. More simultaneous measurements of the equilibrium factor and of the unattached fraction, together with the aerosol concentration and the ventilation rate, are necessary in order to obtain a clear and consistent picture.

138. Information on the equilibrium factors measured in several surveys is given in Table 18. In the Swedish survey [S12], simultaneous measurements of the air exchange rate and of the equilibrium factors were performed; the equilibrium factor was found to increase as the air exchange rate decreased ($F = 0.33$ for $\lambda_s > 0.6$ h⁻¹; $F = 0.51$ for $\lambda_s < 0.3$ h⁻¹). The average values obtained in the surveys listed in Table 18 lie between 0.3 and 0.8, most of the values being on the low side of this range. In this Annex, a value of 0.4 is adopted as the average equilibrium factor for radon...
decay products in dwellings. This is slightly lower than the figure of 0.5 selected in the UNSCEAR 1977 and 1982 Reports [U2, U1].

3. Results of indoor surveys

139. Large-scale surveys, usually involving more than 100 dwellings, have recently been completed or are in progress in several countries (Table 19). The purposes of these large-scale surveys are to study the distribution of population exposures, search for problem areas, or suggest remedial action to reduce elevated natural levels or man-made enhancements. An analysis of the results presented in Table 19 shows that (a) in all cases, the radon and radon decay product concentrations fit as a first approximation, a log-normal distribution. As an example, Figure XXIV shows the histogram of indoor radon concentrations obtained in a survey in Sweden [M30, S8], which conforms reasonably well to a log-normal distribution with a geometric standard deviation of 3.4. The geometric standard deviations reported in Table 19 range from 1.6 to 5.2. The medians of the equilibrium equivalent concentrations of radon range from less than 4 Bq m\(^{-3}\) in Poland [G13] to about 30 Bq m\(^{-3}\) in Sweden [S8] and Switzerland [B4]; in the tail of the distribution, relatively high concentrations of the order of 100 Bq m\(^{-3}\) (EEC) or more were obtained; (b) the highest radon concentrations were caused by a high influx of radon from the soil, a high exhalation rate from building materials, a high radon concentration in tap-water or a combination of the above factors, depending on environmental characteristics and the building design. In houses, a large fraction of the highest radon concentrations could be consistently attributed to high radon entry rates from the soil; in apartments, the influence of building materials was more important; (c) in most of the older surveys radon and/or radon decay products were sampled under conditions leading to high concentrations (basements or unventilated rooms). In more recent surveys, the sampling has been carried out over periods lasting from a few weeks to a year in order to average out the temporal variations; the spatial variations could also be taken into account by installing the measuring devices in two or three locations (living-room, bedroom, basement). Thus, the intention in recent surveys was to attempt to assess the exposures in buildings under realistic as opposed to worst-case conditions; and (d) owing to the large influence of the radon influx from the soil, radon concentrations tended on average to be greater in houses than in apartments. This was clearly shown in the Swedish survey [S8], as well as in the survey from the Federal Republic of Germany [W15], and may explain the relatively low radon concentrations in Poland [G13] and Austria [S16, S17], where a large proportion of the dwellings are apartment buildings.

140. In the UNSCEAR 1982 Report (Annex D, paragraph 203) it was estimated that the mean indoor equilibrium equivalent concentration of radon was 15 Bq m\(^{-3}\) in the temperate regions of the world. The results presented in Table 19, although preliminary for a few surveys, make it possible to reassess this value. The population-weighted mean of the indoor radon concentrations obtained in the nation-wide surveys was calculated assuming a log-normal distribution and a geometric standard deviation of 2.5 for countries that reported only the median concentration. The countries considered in the assessment are in the temperate and high latitudes and represent about 750 million people. The population-weighted mean indoor concentration is found to be 51 Bq m\(^{-3}\) while the arithmetic mean of the country means is 55 Bq m\(^{-3}\). This is in agreement, perhaps as a coincidence, with the mean radon concentration of 50 Bq m\(^{-3}\) obtained

![Figure XXIV. Distribution of indoor radon concentrations in Swedish homes. (M30, S8)](image-url)
for the reference house with a radon entry rate of 50 Bq h\(^{-1}\) m\(^{-3}\) and an air exchange rate of 1 h\(^{-1}\). Using that round value of 50 Bq m\(^{-3}\) and an average equilibrium factor between radon and its decay products of 0.4 yields an average indoor equilibrium equivalent concentration of radon of 20 Bq m\(^{-3}\) in the temperate regions of the world; the geometric standard deviation of the distribution is expected to be about 2.5.

141. There is little information on the indoor radon concentrations in the tropical regions. At Bombay, India, an indoor radon concentration of 19 Bq m\(^{-3}\) and an equilibrium factor of 0.43 are reported as typical for normal areas [M29], while a mean radon concentration of 34 Bq m\(^{-3}\) may be derived from indoor measurements at Hong Kong [T13], which is not thought to be representative of tropical regions. It is safe to assume that the mean indoor radon concentrations in tropical regions are smaller than in temperate latitudes but the data base available is too small to allow a reliable assessment to be made. Possible values of the population-weighted world average of the indoor concentrations of radon gas, taking into account that the population in tropical regions is about half that in temperate latitudes, range from 33 Bq m\(^{-3}\) (assuming that the radon concentration in tropical regions is equal to zero) to 50 Bq m\(^{-3}\) (assuming that the mean radon concentration in tropical regions is the same as that in the temperate zones). It is tentatively assumed in this document that the population-weighted average of the indoor radon concentrations is 40 Bq m\(^{-3}\). The same method can be used for the equilibrium equivalent radon concentration; the range of possible values is from 13 to 20 Bq m\(^{-3}\) and the mean population-weighted world average of the indoor equilibrium equivalent concentrations of radon is tentatively assumed to be 15 Bq m\(^{-3}\).

C. EXPOSURE-DOSE RELATIONSHIPS

1. Inhalation

   (a) Relationships from the UNSCEAR 1982 Report

142. Because of their different physical properties, radon gas and radon decay products are considered separately. Inhaled radon, being a noble gas, is constantly present in the air volume of the lungs at the concentration in air and is partly dissolved in soft tissues. Taking the solubility factor for soft tissues to be 0.4 and assuming that the short-lived decay products decay in the same tissue as radon gas, the following relationship for soft tissues other than the lungs was derived from [12]:

\[
\bar{D}_{\text{soft tissues}} (\text{nGy h}^{-1}) = 0.005 \chi_{\text{Rn,ne}} \text{(Bq m}^{-3}\text{)} \tag{2.19}
\]

In the case of the lungs, in addition to the dissolved radon, the radon content of air in the lungs must be taken into account. Assuming the air volume in the lungs to be 3.2 \(10^{-3}\) m\(^3\) for Reference Man and assuming further that the short-lived decay products will stay in the lungs, the dose rate due to alpha-radiation was obtained as [12]:

\[
\bar{D}_{\text{lung}} (\text{nGy h}^{-1}) = 0.04 \chi_{\text{Rn,ne}} \text{(Bq m}^{-3}\text{)} \tag{2.20}
\]

Taking a quality factor of 20 for alpha-radiation and applying a weighting factor of 0.12 for the lungs and of 0.88 for the other tissues, the effective dose equivalent rate was calculated as

\[
\bar{H}_{\text{eff}} (\text{nSv h}^{-1}) = 0.18 \chi_{\text{Rn,ne}} \text{(Bq m}^{-3}\text{)} \tag{2.21}
\]

143. For the short-lived decay products of radon, use was made of the potential alpha-energy intake by inhalation during time period T, which is given by

\[
I_{\text{pot}} = E/A I_{\text{in}} T \chi_{\text{eq,Rn}} \tag{2.22}
\]

where \(I_{\text{pot}}\) is expressed in joules; \(E/A = 55.4 \times 10^{-10}\) J Bq\(^{-1}\) is the potential alpha energy per unit of inhaled equilibrium equivalent activity; \(I_{\text{in}}\) is the mean breathing rate, taken to be equal to 0.8 m\(^3\) h\(^{-1}\) indoors and 1 m\(^3\) h\(^{-1}\) outdoors; \(T\) is the time period in hours; \(\chi_{\text{eq,Rn}}\) is the equilibrium equivalent concentration of radon daughters in Bq m\(^{-3}\).

144. The dosimetry must consider two target tissues in the lung: the tracheo-bronchial basal cell layer (T-B) and the pulmonary epithelium (P), the latter including the alveolar region and the non-ciliated terminal bronchioles. In the UNSCEAR 1982 Report (Annex D, paragraphs 140-144), the conclusions of an expert group of the Nuclear Energy Agency (NEA) of OECD [N15] were adopted after taking into consideration various dosimetric models [H3, H4, J5, J6, J7]. The Committee indicated that from the dosimetric models, it follows that the conversion factors between the inhaled potential alpha-energy intake \(I_{\text{pot}}\) of the decay product mixture and the committed dose equivalent \(H\) to the tracheo-bronchial basal cell layer and to the pulmonary epithelium show nearly linear relations with the unattached fraction \(f_p\) of the total potential alpha energy of the decay product mixture:

\[
H_{\text{T-B}}/I_{\text{pot}} = a_{\text{T-B}} + b_{\text{T-B}} f_p \tag{2.23}
\]

and

\[
H_P/I_{\text{pot}} = a_P (1 - f_p) \tag{2.24}
\]

where the term \(a\) gives the conversion coefficient for the attached decay products, which depends on the activity median diameter (AMD) of the carrier aerosol. It should be noted that the calculated value of \(I_{\text{pot}}\) applies only to the specified breathing rate.

145. Taking \(f_p = 0.025\) and AMD = 0.2 \(\mu\)m for the indoor environment, the different dosimetric models yield

\[
D_{\text{T-B}}/I_{\text{pot}} = 1.2 - 1.7 \text{ Gy J}^{-1}\]

and

\[
D_P/I_{\text{pot}} = 0.13 - 0.4 \text{ Gy J}^{-1}\]

For the purpose of dose estimation, values of \(D_{\text{T-B}}/I_{\text{pot}} = 1.5 \text{ Gy J}^{-1}\) and \(D_P/I_{\text{pot}} = 0.2 \text{ Gy J}^{-1}\) were selected. Following the ICRP recommendations [12], the Committee decided to split the weighting factor for the total lung and to apply a weighting factor \(w = 0.06\) to each of the two target tissues in the lungs. As the doses to tissues other than the lungs can be
neglected, the effective dose equivalent per unit of inhaled potential alpha-energy intake of \(^{222}\text{Rn}\) decay products was found to be in the range of

\[
H_{\text{eff}}/I_{\text{pot}} = 1 - 3 \text{ Sv J}^{-1}
\]

A reference value of 2.5 Sv J\(^{-1}\) was adopted in the UNSCEAR 1982 Report for indoor exposures.

146. For the exposures in outdoor air, AMD was taken to be 0.1 \(\mu\)m, leading to a dose equivalent per unit intake higher by a factor of 1.5 than those for indoor exposures:

\[
\hat{D}_{T-B}/I_{\text{pot}} = 2.2 \text{ Gy J}^{-1}
\]

\[
\hat{D}_{P}/I_{\text{pot}} = 0.3 \text{ Gy J}^{-1}
\]

147. Assuming steady-state conditions, the dose rates, in nGy h\(^{-1}\), are, for indoor exposures, approximately equal to

\[
\hat{D}_{T-B} = 7 X_{eq,\text{Rn}} \text{ and } \hat{D}_{P} = 0.9 X_{eq,\text{Rn}}
\]

corresponding to effective dose equivalent rates, in nSv h\(^{-1}\), of

\[
H_{\text{eff}} = 9 X_{eq,\text{Rn}}
\]

148. For outdoor exposures, the dose rates, in nGy h\(^{-1}\), are found to be

\[
\hat{D}_{T-B} = 12 X_{eq,\text{Rn}} \text{ and } \hat{D}_{P} = 1.7 X_{eq,\text{Rn}}
\]

while the effective dose equivalent rate, in nSv h\(^{-1}\), is

\[
H_{\text{eff}} = 17 X_{eq,\text{Rn}}
\]

As indicated later (paragraph 153), this is an overestimate.

149. All the dosimetric coefficients given above refer to adult members of the public. Correction factors should be applied for infants and children, to account for the age-dependent change in lung mass and breathing rate. The UNSCEAR 1982 Report [Annex D, paragraph 151] estimated on the basis of the NEA expert report [N15] that the effective dose equivalent for the age group of up to ten years might on the average be a factor of 1.5-2 higher than for adults.

(b) Consideration of other information

150. NCRP [N14] estimated the variability of the bronchial dose from inhalation of radon decay products due to the uncertainties attached to the many physical and biological parameters which are involved in the calculation of absorbed doses per unit concentration of radon decay products. The physical characteristics include the fraction of unattached \(^{218}\text{Po}\) in the atmosphere, the daughter product equilibrium, particle deposition models, the particle size distribution in the atmosphere and the physical dose calculation. Among the biological characteristics are the breathing pattern (including nasal deposition), bronchial morphometry, the mucociliary clearance rate, the location of target cells and the mucus thickness.

151. The dosimetric model used by NCRP to derive the bronchial dose is different from that used in the UNSCEAR 1982 Report but the parameters are the same and the uncertainties should be qualitatively similar. The results obtained by NCRP are given in Table 20 as per cent changes from the nominal annual dose for males. The standard deviation of the annual dose to an individual is estimated to be 120\%, if the various factors other than the radon concentration are unspecified, and less than 100\% that is, better than a factor of 2, if some of the parameters are measured or reasonably well estimated [N14].

152. According to Table 20, the estimate of bronchial dose is very sensitive to the particle size spectrum and to the location of target cells in the tracheo-bronchial tree. The per cent variation in annual dose for a very large shift in the median particle size from an average of 0.125 \(\mu\)m would range from \(+100\%\) for the 0.05 \(\mu\)m particle to \(-20\%\) for the 0.17 \(\mu\)m particle.

153. The values of AMD adopted in the UNSCEAR 1982 Report for the calculation of doses in the respiratory system from inhalation of radon decay products were 0.1 \(\mu\)m and 0.2 \(\mu\)m for outdoor and indoor exposures, respectively. Reported indoor AMDs are 0.125 \(\mu\)m in the United States [G11] and 0.17 \(\mu\)m \((\sigma_g = 3.5)\) in the Federal Republic of Germany [B30, P5]. The doses calculated by the Committee for indoor exposures may thus be slightly underestimated. Regarding the situation outdoors, a significant shift to large particle sizes with a mean AMD of 0.39 \(\mu\)m and a mean \(\sigma_g\) of 2.3 [P5] was observed in the Federal Republic of Germany [B30]; a similar shift had also been measured in the United States [S45]. In another series of measurements in the United States, the AMD was found to range from 0.09 to 0.37 \(\mu\)m with a mean of 0.16 \(\mu\)m [P23]. In the case of outdoor exposures, the doses calculated in the UNSCEAR 1982 Report and shown in paragraph 148 may thus be overestimated by a factor of about 2. If the dose coefficients related to outdoor exposures are divided by two, the resulting values are very similar to the dose coefficients for indoor conditions. In this Annex, the same dosimetric coefficients, in terms of absorbed dose rates per unit equilibrium equivalent radon concentrations, have been used for indoor and outdoor conditions. This implies similar values of the AMD, about 0.2 \(\mu\)m for both indoors and outdoors.

154. The dose factor of 9 nGy h\(^{-1}\) per Bq m\(^{-3}\), as derived in paragraph 147, is based on a median particle size of 0.2 AMD for both indoor and outdoor exposures and on breathing rates of 0.8 m\(^3\) h\(^{-1}\) indoors and 1.0 m\(^3\) h\(^{-1}\) outdoors. Other investigators have developed factors based on other reasonable sizes and rates, as well as other deposition models, without markedly changing the derived dose factors.

155. In the NCRP model [N14], based on the work of Harley and Pasternack [H3], the doses are calculated for the shallow basal cells lying 22 \(\mu\)m below the epithelial surface of the fourth generation of the bronchial tree that was found to experience the maximum deposition density. The deposition model was based on the Weibel lung morphometry [W16] as used by several investigators. The average dose factor for adults was 6 nGy h\(^{-1}\) per WLM or 10 nGy h\(^{-1}\) per Bq m\(^{-3}\) (EEC). Inclusion of later information on morphometry has tended to smooth out the maximum
dose over the entire bronchial tree. Harley and Cohen [H23] measured deposition in a bronchial cast and found it to be very uniform. The dose factor, averaged over all branch airways below the trachea, is 5 mGy per WLM or 8 nGy h⁻¹ per Bq m⁻³ (EEC).

156. Calculations have been performed in Canada of the bronchial dose per unit EEC radon concentration for various indoor aerosol conditions [D14]: the relative range of the bronchial dose per unit EEC radon concentration is from 1 to 5 when indoor aerosol conditions change as a result of seasonal air circulation practices, but is only from 1 to 1.6 when the bronchial dose per unit radon concentration is considered. The bronchial dose rate per unit radon concentration was calculated to range from 2.5-4 nGy h⁻¹ per Bq m⁻³ for adult males.

157. In Belgium, Vanmarcke et al. [V4] measured indoor aerosol characteristics, as well as radon and radon decay product concentrations, under various conditions in several different rooms and calculated the resulting bronchial dose rates per unit radon gas and radon EEC concentration, using the models considered by OECD [N15]. With respect to dose calculations, Vanmarcke et al. [V4], as in [D14], obtained a narrower range when the bronchial dose was related to the measured radon gas concentration rather than to the measured equilibrium equivalent concentration, a typical value being 5 nGy h⁻¹ per Bq m⁻³ of radon gas.

158. James [J19], in a review of all the factors involved in the dosimetry of radon decay products, suggested that it would be appropriate to consider secretory cells as well as basal cells as target cells, implying that the bronchial dose should be calculated for the entire depth of the bronchial epithelium rather than for the basal cells lining the basement membrane alone. This change, combined with the use of a revised deposition model, led to an effective dose equivalent rate per unit EEC radon concentration of 24 nSv h⁻¹ per Bq m⁻³ [J19].

(c) Conclusion

159. The value of bronchial dose rate per unit EEC radon concentration adopted in this Annex for indoor and outdoor radon concentration is 7 nGy h⁻¹ per Bq m⁻³ (see Table 2). If the equilibrium factor is assumed to be 0.4, the bronchial dose rate per unit radon gas concentration is about 3 nGy h⁻¹ per Bq m⁻³, which is in the lower part of the range of values recently published [D4, J19, N22, V4]. The corresponding effective dose equivalent rate is about 10 nSv h⁻¹ per Bq m⁻³ (EEC) including the contribution from the pulmonary region. This is the mean dose in the specified target tissues. It is recognized that the dose factors may vary within a factor of about 3 according to the target cells considered in the dosimetry. Also, recent studies seem to indicate that for indoor conditions, the bronchial dose is better related to the radon gas concentration than to the equilibrium equivalent concentration. These results, however, need confirmation. The dose factors adopted in this Annex are only slightly modified in comparison to those used in the UNSCEAR 1982 Report.

2. Ingestion

160. Ingestion of water containing dissolved radon results in doses in the body tissues from the radon gas and the radon daughters in the water. Much of the dissolved radon is released from water during cooking. The only significant radon intake comes from the drinking of water. The main part of the ingested radon is eliminated from the body very rapidly through the lungs. The largest dose is received by the stomach; estimates of stomach doses per unit activity of radon ingested vary between about 2 and 10 nGy Bq⁻¹ [A3, B2, B3, C20, H7, K7, S15]. In this Annex, an average value of 5 nGy Bq⁻¹ has been adopted.

161. Assuming an average consumption of 0.5 litre of water (taken from the tap, without processing) per day and per person, the annual stomach dose equivalent per unit ²²²Rn concentration in water would be 0.9 nGy per Bq m⁻³. Using a weighting factor of 0.06 and a quality factor of 20 for alpha-radiation, the annual effective dose equivalent per unit radon concentration in water is thus about 1 nSv per Bq m⁻³.

3. Summary

162. Table 21 summarizes the dosimetric coefficients adopted in this Annex for inhalation and ingestion of radon and radon decay products. It is to be noted that these dosimetric coefficients apply only to adults in the general population.

D. DOSES

163. The annual absorbed doses resulting from indoor and outdoor inhalation of radon short-lived decay products have been derived from the estimated mean concentrations and dose coefficients discussed previously, using occupancy factors of 0.8 and 0.2 for indoor and outdoor exposures, respectively. The population-weighted world means of the annual absorbed doses in the lungs and in tracheo-bronchial cells add up to about 100 μGy and 800 μGy, respectively. The corresponding annual effective dose equivalent amounts to approximately 1,100 μSv, the contribution of outdoor exposure being 70 μSv and that of indoor exposure 1,000 μSv.

164. It should be stressed that these values apply to adults in the general public and are averaged over large population groups. Very high individual doses can be obtained, especially from indoor exposures at the tail of the log-normal distribution. The situations in which these elevated exposures arise have been discussed in this section.

165. A refined methodology could distinguish the indoor concentrations in dwellings from the indoor concentrations in working places and public buildings; it could also take into account the daily variations of the indoor concentrations and of the occupancy factors.
166. Some information is available on the dependence of indoor radon concentration upon the type of building (see, for example, Figure XXV [W15]). Further surveys of this type should be encouraged.

167. There is abundant information on the daily variation of indoor radon concentrations, which could be reviewed and used in the refined methodology. Figure XXVI [C16] gives an example of measured diurnal variation of indoor radon concentrations.

168. A survey of diurnal variation of indoor and outdoor occupancy factors has been published in the United Kingdom [B23]; it is presented in Figure XXVII. Occupancy factors reported for populations in China [B24] are similar to those obtained in the United Kingdom.

169. Information of the type presented in Figures XXV, XXVI and XXVII can be combined to estimate refined annual absorbed doses from inhalation of...
radon decay products. Further refinements, which could not be implemented at present because of insufficient information, would take into account the age distribution of the populations exposed, urban versus rural population distribution, as well as seasonal variations and the latitudinal distribution of the radon concentrations.

III. INDUSTRIAL ACTIVITIES

170. The industrial processes considered are those that bring to the surface of the earth materials containing above average concentrations of the natural radionuclides, such as geothermal work or phosphate mining, and those which treat material containing average or above average amounts of the natural radionuclides and concentrate them in one or more products or by-products, such as coal burning or the production of fertilizer from phosphate rock. In these industrial processes, the hazard from radiation is generally small compared to that from other chemical substances; therefore, radiation is not systematically monitored. Assessment of such exposures is usually based on sketchy information derived from isolated surveys. The only assessment at the national scale that was available to the Committee is that of the Netherlands [P26]. In this, the naturally occurring radioactive materials discharged to the environment by non-nuclear industries in the Netherlands was estimated using a crude model based on radionuclide concentrations in raw materials, mass flow, and information on the technologies used in industrial processes. The annual effective dose equivalents received by members of the public in the Netherlands were estimated to be about 40 μSv on average, and about 0.7 mSv for individuals of the critical group consisting of consumers of large amounts of fish [P26].

171. This chapter reviews the information available world-wide on radiation exposures from industrial activities, classified into: (a) combustion of coal; (b) other energy production; (c) use of phosphate rock; and (d) metal mining and processing. When possible, both occupational exposures and exposures of members of the public are considered. Collective effective dose equivalent commitments resulting from atmospheric discharges of radioactive materials are estimated using the crude models described in the UNSCEAR 1982 Report ([U1], Annex C, paragraphs 24-27).

A. ENERGY PRODUCTION FROM COAL

172. Coal, like most materials found in nature, contains trace quantities of 40K and of 238U, 232Th and their decay products. By burning coal, the activities of these naturally occurring radionuclides are redistributed from underground into the biosphere. Expressed in coal equivalent for energy purposes, the world production of coal was 3.1 × 10^13 kg in 1985 [U15], the main producers being China, the USSR and the United States. Table 22 presents the amounts of coal produced in various countries and the measured activity mass concentrations of natural radionuclides.

173. Although the reported activity mass concentrations range over two or three orders of magnitude, the averages from the various countries are in fairly good agreement. Figure XXVIII shows, for example, the 238U levels measured in 800 samples of coal mined in the United States [W12]; they range from 1 to 1,000 Bq kg⁻¹. In this Annex, as in the UNSCEAR 1982 Report, it will be assumed that the average activity mass concentrations of 40K, 238U and 232Th in coal are 50, 20 and 20 Bq kg⁻¹, respectively, and that the decay products of 238U and 232Th are in radioactive equilibrium with their precursor.

174. The uses of coal in OECD countries are listed in Table 23, together with the amounts involved in
several countries [06]. Most of the coal is used in electric power stations and in coke ovens; even though the use of coal for space heating is not as important as it was years ago, it is still significant in some countries. It will be assumed that, on a world-wide basis, 70% of the coal produced is burned in electric power stations, 20% in coke ovens and 10% in houses.

175. Radiation exposures occur throughout the coal fuel cycle, which consists of (a) coal mining; (b) use of coal; and (c) use of fly ash. Occupational exposures and exposures of members of the public are estimated, in so far as data are available. It should be emphasized, however, that the contribution of radiation hazards to the overall health and environmental impact resulting from the exploitation of coal is generally estimated to be quite small (see, for example, [B39], [U16] and [M17]).

1. Coal mining

176. Coal miners are exposed to coal dust and the decay products of radon and thoron. Coal mines are usually well ventilated to keep the methane and dust levels low; consequently, the radon concentrations are also relatively low. Table 24 summarizes published measurements from several countries. Radon concentrations in coal mines are comparable to those observed in dwellings. From measurements in British coal mines, a maximum individual effective dose equivalent of 1 mSv per year, and an upper limit for the normalized collective effective dose equivalent of 3 man Sv per GW a have been calculated [C14, R4]. Unquantified additional exposures may result from the activity present in mine water [T8].

177. The global production of electrical energy by coal burning was about 600 GW a in 1980, resulting in an upper estimate of the total collective effective dose equivalent to coal miners of 2,000 man Sv. This figure is taken to represent the collective effective dose equivalent for the world for one year of mining coal, including the mining of coal for purposes other than electrical energy production.

178. Members of the public are exposed to the radon activity present in the exhaust air of coal mines. There are currently no measured data on emission of radon from coal mines. It can be assumed, as in [S32], that 20% of the radon contained in the ore is released during the mining process and that the radon release rate from the exposed ore surface is a factor of 2 greater than that resulting from mining. Taking the mass of coal required to produce an electrical energy of 1 GW a to be 3.10^9 kg and a mean radon concentration in coal of 20 Bq kg^{-1}, the normalized radon release from coal mines would be 36 GBq per GW a. Another method consists in scaling the radon releases from coal mines to the radon releases from uranium mines, taking into account an average factor of 1,000 between the average activities of ^{226}Ra per unit mass of uranium ore and of coal [J15]. This method leads to a normalized radon release from coal mines of 600 GBq per GW a. The normalized collective effective dose equivalent commitments corresponding to the two values of radon releases are 6 \times 10^{-4} and 10^{-2} man Sv per GW a.

179. Using the two methods described in the previous paragraph, the activity of radon released per year from coal mining all over the world is estimated to be 30 TBq and 800 TBq, resulting in collective effective dose equivalent commitments of about 0.5 and 10 man Sv, respectively. Although differing by a factor of 20, the latter values are small compared to other radiation exposures from the coal fuel cycle and do not deserve further investigation.

2. Use of coal

180. The uses of coal that are considered here are electrical energy production, carbonization and space heating in dwellings.

(a) Coal-fired power plants

181. In the production of electric power, coal is burned in furnaces operating at temperatures of up to 1,700°C. About 3 \times 10^9 kg of coal is required to produce 1 GW a of electrical energy. In the combustion process, most of the mineral matter in the coal is fused into a vitrified ash. A portion of the heavier ash, together with incompletely burned organic matter, drops to the bottom of the furnace as bottom ash or slag. The lighter fly ash, however, is carried through the boiler, together with the hot flue gases and any volatilized mineral compounds, to the stack, where, depending on the efficiency of emission control devices, most is collected while the rest (escaping fly ash) is released to the atmosphere. Table 25 presents the ash content of coal burned in the United Kingdom in power stations and for other uses, together with the amounts of ash produced; it is clear that coal combustion in coal-fired power plants accounts for the bulk of the total ash production. The corresponding information on the atmospheric emissions of particulates (Table 26), however, indicates that the power stations are a small contributor to the total emissions of particulates, implying that the efficiency of the emission control devices used in power plants is rather high.

182. The radionuclides included in the non-combustible mineral matter of coal are partitioned between the bottom ash and fly ash, except for the gases and volatilized minerals, which are incorporated directly into the flue gases. In large power stations, there is about 20% bottom ash and, therefore, 80% fly ash. Owing mainly to the elimination of the organic component of the coal, there is approximately an order of magnitude enhancement of the activity concentrations from coal to ash. Consequently, the natural radionuclide concentrations in ash and slag from coal-fired power stations are significantly higher than the corresponding concentrations in the earth's crust. The arithmetic averages of the reported activity mass concentrations in escaping fly ash are 265 Bq kg^{-1} for ^{40}K, 200 Bq kg^{-1} for ^{238}U, 240 Bq kg^{-1} for ^{226}Ra, 930 Bq kg^{-1} for ^{210}Pb, 1,700 Bq kg^{-1} for ^{210}Po, 70 Bq kg^{-1} for ^{222}Th, 110 Bq kg^{-1} for ^{228}Th and 130 Bq kg^{-1} for
$^{226}$Ra ([U1], Annex C, paragraph 11). There is an enrichment by a factor of about 3 for $^{210}$Pb and 5 for $^{210}$Po in relation to the rest of the $^{238}$U decay series. This is probably because of the volatilization of $^{210}$Pb and $^{210}$Po during the combustion and a condensation of these radionuclides down the flue line on the finer fly-ash particles.

183. The activities of natural radionuclides discharged to the atmosphere from a power plant per unit electrical energy produced depend on a number of factors such as the activity concentration in coal, the ash content of the coal, the temperature of combustion, the partitioning between bottom ash and fly ash, and the efficiency of the filtering system. Marked differences should therefore be expected between the measured activities discharged per unit energy produced from different power plants. Most of the available information on the normalized atmospheric discharges is presented in Table 27. Apparently, there are two types of coal-fired power plants used throughout the world: older ones, which release about 10% of the fly ash produced; and more modern ones, which are equipped with sophisticated retention devices and, as a result, release only about 0.5% of the fly ash. Table 27 presents estimates of normalized discharged activities for such older and more modern plants. They have been derived on the following assumptions: the combustion of 3 $10^{6}$ kg of coal is required to produce an electrical energy of 1 GW a; fly ash accounts for 80% of the total ash production; the activity mass concentration in coal is 20 Bq kg$^{-1}$ for the radionuclides of the $^{238}$U and of the $^{232}$Th decay series; and there is an enrichment factor of 3 for $^{210}$Pb and $^{210}$Po. Releases of $^{222}$Rn and of $^{220}$Rn must be estimated separately because radon is not collected by the particulate control devices. The activities of $^{222}$Rn and of $^{220}$Rn released per GW a have been assessed at 60 GBq on the basis of the conservative assumption that all of the radon activity contained in the coal is discharged to the atmosphere.

184. The main pathways through which the populations living around coal-fired power plants are exposed to enhanced levels of natural radionuclides are inhalation during the cloud passage and external irradiation and ingestion following deposition of activity on the ground. The collective effective dose equivalent commitments due to those pathways, normalized to the generation of 1 GW a of electrical energy, have been estimated using the crude models described in the UNSCEAR 1982 Report ([U1], Annex C, paragraphs 24-27). Tables 28 and 29 present the results obtained for the reference older and more modern coal-fired power stations, respectively, they amount to about 6 and 0.3 man Sv per GW a. Assuming that, on a worldwide scale, two thirds of the electrical energy provided by coal-fired power stations is due to old plants (and one third to modern plants), the average normalized collective effective dose equivalent commitment is approximately 4 man Sv per GW a, while the overall figure for the world for one year of practice is about 2,000 man Sv.

185. The release of $^{14}$C-free carbon dioxide emitted by the coal-fired power plants will have the effect of diluting the natural concentration of $^{14}$C in the biosphere. The reduction in the natural $^{14}$C collective effective dose equivalent commitment resulting from the release of activity-free carbon dioxide is estimated to be about 60 man Sv per GW a assuming that: (a) the combustion of about 3 Tg of coal is required to produce an electrical energy of 1 GW a; (b) the coal contains 85% carbon by mass; (c) the natural activity mass concentration of $^{14}$C in coal is approximately 226 Bq kg$^{-1}$; and (d) the collective effective dose equivalent commitment per unit release is 110 man Sv per TBq. This reduction in the natural effective dose equivalent commitment would be over thousands of years.

186. Annual individual effective dose equivalents resulting from inhalation during the cloud passage have been estimated. Assuming an effective stack height of 100 m and a uniform wind rose, the annual average of the ground-level air concentration per unit release rate is estimated to be about 4 $10^{-8}$ Bq m$^{-3}$ per Bq s$^{-1}$ at approximately 1 km from the stack. For an individual of the critical group living near a coal-fired power plant producing annually 1 GW a of electrical energy, the resulting annual effective dose equivalent would be about 1 $\mu$Sv for a modern plant and about 20 $\mu$Sv for an old plant. The annual effective dose equivalents from ingestion and external irradiation would be of about the same magnitude after 20-30 years of operation. Calculated doses from ingestion are very sensitive to the assumptions made about food-chain transfers. Annual individual doses are, in any case, small; it is worth noting that recent measurements in the environment surrounding coal-fired power plants show no statistically significant increases in the activity concentrations of natural radionuclides in samples of air [T7], precipitation [T7], soil [B15, S30, T7], vegetation [S30, T7] or cattle organs [B16, S30, S31] that could be attributed to the operation of power stations.

187. Only atmospheric discharges have been considered so far, as coal ash is not usually directly discharged into the aquatic environment. It may be stored in ash ponds, from where some activity may leach into the aquifer. Ground water may then carry the leachates into the nearest river. The leachability of the ash is quite low [S27], however, so aquatic discharges are not likely to pose a radiation problem, at least in the short term.

188. Radiation exposures of workers in coal-fired power plants are thought to be mainly caused by inhalation of airborne fly ash. Effective dose equivalents of 150 $\mu$Sv per year have been estimated in the United Kingdom for the most exposed groups of power station workers on the basis of exposure to a concentration of respirable fly ash in air of 0.5 mg m$^{-3}$ [C14]. In Yugoslavia, significant concentrations of $^{210}$Pb in the urine of individuals working in a coal-fired power plant burning high activity coal have been reported [B12, B22].

189. Values of collective effective dose equivalent received by workers in coal-fired power plants are not available. An upper estimate can be derived from the
individual effective dose equivalent of 150 μSv per year calculated in the United Kingdom, assuming that a labour force of 500 persons is required to produce an electrical energy of 1 GW a per year. The normalized annual collective effective dose equivalent would be at most 0.1 man Sv per GW a. The global production of electrical energy by coal burning was about 600 GW a in 1980, resulting in an upper estimate of the total collective effective dose equivalent to workers in coal-fired power plants of 60 man Sv.

(b) Domestic use

190. Another major use of coal is domestic cooking and heating. No information has been found in the literature on the environmental discharges of natural radionuclides from this source. The use of coal for cooking and heating in private homes may, however, be estimated to result in high collective dose commitments since chimneys are generally low and not equipped with ash removal systems and the population densities around sources of emission are generally high. Figure XXIX [U16] presents trends in urban smoke concentrations in the United Kingdom and shows that domestic sources, which have decreased in importance since 1960, still remain substantial. It is estimated that in a simple open fire burning coal, some 3-4% of the coal is emitted as tarry smoke while in smoke-reducing appliances, which have only been marketed since the early 1970s, only 0.3-0.6% of the coal burned is emitted as smoke [U16]. Smoke is a mixture of soot, tarry matter and inorganic substances, produced when bituminous coal is incompletely burned. Measured activity mass concentrations of natural radionuclides in smoke have not been found, but they can be expected to be between those in coal and ash (without enrichment) and are likely to be closer to those in coal. Assuming that the activity mass concentrations in smoke are equal to those in coal and that 3.5% of the coal is emitted as smoke, the annual world-wide atmospheric releases caused by domestic burning of coal are estimated to be 0.7 TBq of 40K and 0.3 TBq of each of the radionuclides of the 235U and 222Th series (radon and thoron excepted); these figures become 20 times greater if it is assumed that the activity mass concentrations in smoke are equal to those in ash and that the coal burned has a 5% ash content. Taking the average population densities around the houses to be 10^3 km^-2 leads to collective effective dose equivalent commitments resulting from a yearly world wide use of coal in the range of 2,000-40,000 man Sv. This estimate is highly uncertain, as it is not supported by any discharge or environmental data.

(c) Coke ovens

191. Coal is also used in vast amounts in coke ovens; however, relevant information on the releases of natural radionuclides from coke ovens has not been found.

3. Use of fuel ash

192. Large quantities of coal ash (fly ash and bottom ash combined) are produced each year throughout the world. The global production of coal equivalent in 1985 was 3.1 x 10^{12} kg (Table 22); assuming that coal burned in power stations represents 70% of the total

![Figure XXIX. Smoke: trends in emissions and average urban concentrations in the United Kingdom. [U16]](image-url)
consumption and that its ash content is 13%, about 280 million tonnes of coal ash are produced annually in coal-fired power stations. Taking the average activity mass concentrations of natural radionuclides in coal to be 50 Bq kg⁻¹ for ⁴⁰K and 20 Bq kg⁻¹ for ²³⁸U and for ²³²Th, the average activity mass concentrations of ⁴⁰K, ²³⁸U and ²³²Th in coal ash are estimated to be 400, 150 and 150 Bq kg⁻¹, respectively. Compared with normal soil, the average activity mass concentration of ⁴⁰K is similar, but those of ²³⁸U and ²³²Th are six times higher.

193. Coal ash is used in a variety of applications, the largest of which is the manufacture of cement and concrete. It is also used as a road stabilizer, as road fill, in asphalt mix and as fertilizer. Data on the various uses of coal ash in several countries have been reported [G20]. It can be estimated that about 5% of the total ash production from coal-burning power stations is used for constructing dwellings: this represents an annual usage of 14 million tonnes.

194. The use of coal ash in building materials may result in changes in the indoor doses owing to external irradiation and to inhalation of radon decay products. With respect to external irradiation, Siotis and Wrixon [S5], assuming that the use of concrete containing fly ash for constructing dwellings would result in an additional activity mass concentration of ²²⁶Ra in concrete of 13 Bq kg⁻¹, calculated that the additional effective dose equivalent from indoor exposure would amount to 20 μSv per year. Stranden [S28], on the basis of additional activity mass concentrations of 10 Bq kg⁻¹ of ²²⁶Ra and 8 Bq kg⁻¹ of ²²⁸Th, estimated that the additional absorbed dose in air would be 90 μGy per year in concrete houses and 40 μGy per year in wooden houses. These figures would be reduced to 70 and 30 μSv for concrete and wooden houses, respectively, if the conversion coefficient from absorbed dose in air to effective dose equivalent were taken to be 0.7.

195. There are conflicting views on the impact of the use of fly ash on the dose from inhalation of radon decay products. Siotis and Wrixon [S5] measured the radon exhalation rates of concrete blocks made by a cement manufacturer under controlled conditions using cement containing varying proportions of fly ash of high ²²⁶Ra activity mass concentration (330 and 800 Bq kg⁻¹). The concrete blocks containing fly ash were found to present radon exhalation rates higher than those of concrete blocks without fly ash. Siotis and Wrixon [S5] estimated, on the basis of these measurements, that the additional radon concentration in a model dwelling would be 1.5 Bq m⁻³, resulting in an effective dose equivalent due to indoor exposure to radon decay products of about 40 μSv per year. Radon concentrations in Polish dwellings built with concrete blocks made almost entirely of fly ash were found to be higher than those in dwellings built with concrete blocks containing no fly ash [B14]. Stranden [S28], however, observed lower radon exhalation rates from concrete blocks containing 5% fly ash than from ordinary concrete blocks; the two types of blocks had been especially constructed for the experiment and differed only in the inclusion of fly ash. The resulting reductions in the radon concentrations in model dwellings have been calculated by Stranden [S28] to be 3 Bq m⁻³ in a wooden dwelling and 10 Bq m⁻³ in a concrete dwelling. The corresponding reduction in effective dose equivalent from indoor exposure to radon decay products would be 90 μSv per year in a wooden building and 300 μSv per year in a concrete dwelling. In other controlled studies [U6, V3], the substitution of varying amounts of ordinary cement with fly ash cement yielded no significant change in the radon exhalation rate from the concrete obtained, in comparison with ordinary concrete.

196. The conflicting evidence may be attributed to differences in the properties of the components used in concrete, their relative activity mass concentrations of ²²⁶Ra or the manufacturing process. It seems that for a given activity mass concentration of ²²⁶Ra, the radon exhalation rate of fly-ash is usually lower than that of concrete, so that enhanced indoor exposures to radon decay products may arise only when fly ash with a high activity mass concentration of ²²⁶Ra is mixed with concrete.

197. In this Annex, the use of fly ash in building materials is assumed to result only in additional exposures from external irradiation. Annual additional effective dose equivalents of 30 μSv in wooden dwellings and 70 μSv in concrete dwellings are adopted as representative values on a world-wide basis. Taking the amount of fly ash concrete to be 1.3 tonnes in a wooden house and 4 tonnes in a concrete building (as in [S28]), the additional effective dose equivalents normalized per unit mass of fly ash concrete are found to be 23 and 18 μSv a⁻¹ per tonne for wooden and concrete dwellings, respectively. Using an intermediate value of 20 μSv a⁻¹ per tonne and assuming that an average of four persons live in each house and that the lifetime of the house is 50 years, the collective effective dose equivalent commitment arising from external irradiation attributable to the annual use of fly ash for constructing the dwellings is estimated to be about 5 10⁻⁴.

198. The use of fly ash as a component of cement and concrete for constructing dwellings represents only a small fraction of the total use of coal ash. The Committee has not found any published information on doses arising from other uses of fly ash (as a road stabilizer, in asphalt mix, as fertilizer, etc.).

199. The large fraction of coal ash that does not find a commercial application is usually dumped in the vicinity of the power plant. Two disposal methods are used. Ash mixed with water in a ratio of 2 to 3 is piped as slurry to lagoons created by adding walls to or otherwise modifying existing topographic features. When a lagoon is filled, it is allowed to drain and, if necessary, another lagoon is created on top. For dry ash dumps, the ash is conditioned with water to reduce dust and transported to the dumping site. The choice of disposal method depends on economic and environmental factors [U6]. When the dumping is finished, most dry ash dumps are covered by topsoil and converted into areas for agricultural or recreational use.
200. The main potential radiation hazards from ash disposal arise from resuspension and from contamination of surface and underground water either as a result of leaching, i.e., as certain compounds are washed out of the ash by percolating rain water, or as a consequence of the physical erosion of the ash surface during run-off. Dose assessments taking into account these processes are lacking.

4. Summary

201. Radiation exposures resulting from the various steps in the coal fuel cycle are summarized in Table 30. The exposures are expressed in terms of collective effective dose equivalent commitments per year of practice around the world. The highest exposures seem to stem from the use of coal in homes and the use of fly ash in concrete. It should be borne in mind that these estimates are fraught with large uncertainties that are difficult to quantify.

B. OTHER ENERGY PRODUCTION

202. The other types of energy production considered in this section are geothermal energy and combustion of oil and natural gas.

1. Geothermal energy

203. Geothermal energy is produced in Iceland, Italy, Japan, New Zealand, the USSR and the United States. In 1980, it amounted to only 1.5 GW a of electrical energy production, but its relative importance may grow in the future. Geothermal energy makes use of hot steam or water derived from high-temperature rocks deep inside the earth. Most of the activity found in geothermal fluids is due to the uranium decay chain. Isotopes of solid elements may cause problems regarding water pollution or land disposal but only radon, which is released into the atmosphere when the water or steam contacts the air, is considered here. From measurements of radon in geothermal fluids in Italy [G18, M15], the annual releases of radon have been estimated to be 110 TBq from the 400 MW Larderello plant, 7.0 TBq from the 15 MW plant at Piancastagnaio and 1.5 TBq from the 3 MW plant at Bagnore. The annual release of radon from the 11 units of the Geysers Power Plant in the United States, which has a net capacity of 502 MW of electrical energy, was assessed to be 21 TBq [A16]. These figures point to an average atmospheric discharge per unit energy generated of about 150 TBq per GW a. The corresponding collective effective dose equivalent commitment per unit energy generated is estimated to be about 2 man Sv per GW year if it is assumed that the equilibrium factor between radon and its short-lived decay products is 0.8, the population density around the plant is 100 km², the effective dose equivalent per unit activity inhaled is 1.1 $10^{-8}$ Sv Bq⁻¹ and that indoor and outdoor concentrations are the same. The annual world-wide production of geothermal energy would yield a collective effective dose equivalent commitment of approximately 3 man Sv.

204. Annual individual effective dose equivalents resulting from inhalation of short-lived decay products of radon is estimated to be of the order of 10 $\mu$Sv at 1 km from the stack of a 100 MW geothermal plant. The average additional radon concentration would be about 0.1 Bq m⁻³, compared with a background concentration of 1-10 Bq m⁻³. Measurements conducted near the Larderello plant in Italy and near other geothermal power plants in New Zealand and the United States failed to distinguish between the contribution of the radon activity from the plant and that from background [A16, G18, S59, W20].

2. Oil-fired power plants

205. As shown in Table 31, oil has a large number of applications, the most important being road transport, generation of electric power and domestic heating. Approximately 3 $10^{12}$ kg of crude petroleum was produced in the world in 1980. In power plants, about 2 $10^9$ kg of oil is needed to produce 1 GW a of electrical energy. As the ash content of oil is very low, oil-fired power plants are usually not equipped with efficient ash removal systems. From measurements in two oil-fired power plants in France, normalized atmospheric discharges of about 200, 300, 150 and 1,000 MBq per GW a for $^{226}U$, $^{228}Ra$, $^{210}Th$ and $^{40}K$, respectively, have been estimated [A8]. These are very similar to the normalized discharges of coal-fired power plants fitted with efficient aerosol control devices. Measurements of activity mass concentrations of $^{212}Th$, $^{228}Ra$, $^{210}Pb$ and $^{40}K$ in fuel ash from oil-burning power stations of the USSR led to the same conclusion [B13]. The collective effective dose equivalent commitment per unit electrical energy generated has been found to be about 0.5 man Sv per GW a. Assuming that 15% of the world-wide production of crude petroleum is burned in electric power plants, the resulting annual collective effective dose equivalent commitment would be about 100 man Sv. The individual effective dose equivalent to a member of the critical group, calculated in the same way as for a coal-fired power plant, would be about 1 $\mu$Sv.

206. Information on radiation exposures of workers in the oil industry is scarce. Kolb and Wojcik [K22, K23] measured a median $^{226}Ra$ concentration of 4,500 Bq m⁻³ in thermal brine, which is raised to the surface together with oil. This brine can be used for the extraction of oil from its deposits without any radiation problem as long as the radium activity remains in solution. Salt from oversaturated brine is often precipitated after expansion, however, and scale is formed on the inner walls of tubing, pumps and separation and storage tanks [K23, S60]. Such deposits can be detected with common radiation protection instruments; activity mass concentrations of up to 10⁶ Bq kg⁻¹ of $^{226}Ra$ and $^{228}Ra$ have been detected in some samples. Under normal conditions of operation, the annual effective dose equivalent for maintenance personnel resulting from external radiation from such sources is estimated to be less than 5 mSv [K23].

207. Oil shale is currently being considered for the production of synthetic oil that could be burned in
power plants. Oil shale is a sedimentary rock containing kerogen, an organic substance originating from micro-organisms that accumulated in what were once large shallow lakes. When the rock is heated to a temperature of at least 400°C to bring about pyrolysis of the kerogen, synthetic shale oil is produced. The production of shale oil involves the processing of vast amounts of material. Retorting 1 tonne of shale can yield 110 litres of retort water, 0.8 tonne of spent shale and 1-40 m³ of retort gases [F6].

208. In the rich shale deposits of Colorado, Wyoming and Utah, known as the Green River formation, as well as in Estonian shale, measured radionuclide concentrations of raw oil shale (about 60 Bq kg⁻¹ of ²³⁵U, 25 Bq kg⁻¹ of ²³²Th, 500 Bq kg⁻¹ of ⁴⁰K) correspond fairly closely to those observed in typical soils [G19]. Antrim shales from the eastern United States and Moroccan shales show higher uranium concentrations (about 350 Bq kg⁻¹) [G19]. After retorting, nearly all of the activity content is found in the spent shale, with a marked increase with decreasing particle size for most radionuclides. Average concentrations in the respirable fraction of spent shale from the Green River formation are about 150 Bq kg⁻¹ of ²³⁵U, 130 Bq kg⁻¹ of ²³⁸Ra, 220 Bq kg⁻¹ of ²¹⁰Pb, 40 Bq kg⁻¹ of ²¹²Th and 740 Bq kg⁻¹ of ⁴⁰K [G19]. Atmospheric discharges from an oil shale plant producing enough oil to generate 1 GW a of electrical energy have been estimated to be about 290 MBq of ²³⁵U, 250 MBq of ²¹⁰Pb, 430 MBq of ²¹⁰Ra, 90 MBq of ²²⁲Th and 1,500 MBq of ⁴⁰K [G19]. This is again very similar to the normalized discharges from a coal-fired power plant equipped with an efficient ash removal system. The collective dose equivalent commitment per unit electrical energy generated would be about 0.5 mSv per GW a.

3. Natural gas

209. In 1980, the world-wide production of natural gas amounted to about 10¹² m³. Like oil, natural gas has many fields of application. Its uses in OECD countries are summarized in Table 32, the major uses being domestic heating, generation of electric power and as a source of heat in various industries. As discussed previously in this Annex, radon concentrations in natural gas may vary widely around a typical value of 1,000 Bq m⁻³. Since about 2 x 10⁸ m³ of natural gas must be burned in order to produce 1 GW a of electrical energy, the corresponding radon emission is approximately 2 TBq.

210. The corresponding collective effective dose equivalent commitment per unit energy generated is estimated to be about 0.03 mSv per GW a. The assumptions for this estimate are that the equilibrium factor between radon and its short-lived decay products is 0.8, the population density around the plant is 100 km⁻², the effective dose equivalent per unit activity is 1.1 x 10⁻⁸ Sv Bq⁻¹ and indoor and outdoor concentrations are the same. Assuming that 15% of the world production of natural gas is burned in electric power plants, the resulting collective effective dose equivalent commitment per year of practice would be about 3 mSv.

211. The radon concentrations around a gas-fired power plant are expected to be, in most cases, undistinguishable from background; the individual doses are expected to be trivial.

4. Peat

212. Peat is burned for energy production in several countries, notably in Finland and Sweden [E9, E10, M31]. Natural radionuclides are carried into the peat bogs by flowing surface and ground water and then adsorbed to peat matter. In Sweden, the normal ²³⁵U activity mass concentration in dried peat is about 40 Bq kg⁻¹, but some samples have shown concentrations up to 500 Bq kg⁻¹; in one extreme case, the ²³⁵U concentration was more than 10,000 Bq kg⁻¹ [E9]. In Finland, samples of peat used as fuel in a power plant showed concentrations of 16 Bq kg⁻¹ of ²³⁵U, 30 Bq kg⁻¹ of ²¹⁰Pb, 5.3 Bq kg⁻¹ of ²¹⁸Ra and 28 Bq kg⁻¹ of ⁴⁰K [M31]. Since the ash content of peat normally lies between 3% and 8% [M31], the concentrations of natural radionuclides in peat ash are about 20 times those in peat.

213. There is little information on the environmental discharges of natural radionuclides from peat power plants. Assuming that 5 x 10¹² kg of peat is needed to produce 1 GW a of electrical energy and using an average ²³⁵U concentration in peat of 40 Bq kg⁻¹ and a dust control efficiency of 99%, as well as the general assumptions used for coal-fired power plants, the normalized collective effective dose equivalent commitment is tentatively estimated to be 2 man Sv per GW a. In the long term, storage and disposal of uranium-rich peat ash may have the greatest impact [E9].

5. Summary

214. Radiation exposures attributable to natural radionuclide discharges from various systems of electricity production are summarized in Table 33. The exposures are expressed in terms of collective effective dose equivalent commitments for one year of practice throughout the world and are normalized to the generation of 1 GW a of electrical energy. The highest radiation exposures result from the use of coal.

C. USE OF PHOSPHATE ROCK

215. Phosphate rock is the starting material for the production of all phosphate products; it is the main source of phosphorus for fertilizers. The world production of phosphate rock was about 130 million tonnes in 1982 [F9], the main producers being China, Morocco, the USSR and the United States. There are three types of phosphate rock: (a) phosphate rock of sedimentary origin: this type, which covers 85% of current needs for phosphorus, comes from deposits spread almost throughout the world, but especially in Morocco and Florida (United States) [111]; (b) phosphate rock of volcanic origin: the principal deposit of this type is found in the Kola peninsula (USSR); (c) phosphate rock of biological origin: the accumulated
droppings of marine birds have given rise to deposits of guano, from which the leached phosphoric acid has recombined with calcium from the underlying rock to give tricalcium phosphate.

216. Activity mass concentrations of natural radioisotopes in phosphate rock were reviewed by UNSCEAR in its 1977 and 1982 Reports [U2, U1]. Activity mass concentrations of $^{237}$Th and $^{40}$K in phosphate rocks of all types are similar to those observed normally in soil, whereas concentrations of $^{238}$U and its decay products tend to be elevated in phosphate deposits of sedimentary origin. A typical concentration of $^{238}$U in sedimentary phosphate deposits is 1,500 Bq kg$^{-1}$. North American phosphate presents the highest concentrations of $^{238}$U, followed by north African phosphate. Uranium-238 and its decay products are generally found in close radioactive equilibrium in phosphate ore.

217. Exposures of members of the public result from the following: (a) effluent discharges of radioactive materials of the $^{238}$U decay series into the environment from phosphate rock mining and processing; (b) use of phosphate fertilizers; and (c) use of by-products and wastes.

218. Occupational exposures mainly occur during mining, processing and transportation of phosphate rock, as well as during transportation and utilization of phosphate fertilizers.

1. Effluent discharges during phosphate industrial operations

219. Phosphate industrial operations can be divided into mining and milling of phosphate ore, phosphate product manufacture by the wet process (in wet-process phosphoric acid plants) and phosphate product manufacture by the thermal process (in elemental phosphorus plants).

220. The technique used to mine phosphate ores varies from area to area depending on the type of deposit. In Florida phosphate ore is mined using the strip-mining technique, which involves stripping the overburden and mining the matrix with draglines creating mine cuts from 15 to 20 m in depth [R5]. Overburden is stored near mine cuts and used as backfill as subsequent cuts are made. Structures later built on reclaimed land may exhibit elevated radon concentrations, as discussed later in this Annex.

221. After mining, beneficiation, which is a physical separation process using screen flotation to remove non-phosphate components, is applied to phosphate ores not rich enough in $\text{P}_2\text{O}_5$. Exposures resulting from mining and beneficiation have not been assessed.

222. After phosphate rock has been mined and beneficiated, it is usually dried and ground to a uniform particle size to facilitate processing. Wet process plants produce phosphoric acid, the starting material for ammonium phosphate and triple superphosphate fertilizers; in that process, phosphogypsum is produced as waste or by-product. Thermal process plants produce elemental phosphorus, which is in turn used primarily for the production of high-grade phosphoric acid, phosphate-based detergents, and organic chemicals. Waste and by-products of the thermal process are slag and ferrophosphorus.

223. The drying and grinding operations that follow phosphate rock mining and beneficiation produce significant quantities of particulate material [U9]. Very limited data are available on actual field measurements of radioactive materials from drying and grinding operations. Measurements made by the United States Environmental Protection Agency (EPA) [P16] were summarized in the UNSCEAR 1982 Report [U1]. EPA has since established the parameters of a reference phosphate rock drying and grinding plant [U9], which it used to calculate the emissions of radioactive materials into the atmosphere (see Table 34); they correspond to a particulate emission rate of about 0.1 kg per tonne of phosphate rock throughput and to radioactive equilibrium of $^{238}$U with its solid decay products. The atmospheric release of $^{222}$Rn has not been estimated by EPA; assuming an activity mass concentration of $^{226}$Ra in phosphate ore of 1,500 Bq kg$^{-1}$ and, conservatively, a 100% release of $^{222}$Rn, would yield 1.5 MBq per tonne of phosphate rock.

224. Some phosphate rock must be calcined before it can be processed. The need for calcining is determined primarily by the amount of organic material in the beneficiated rock. In the calcining operation, the rock is heated to about 800°C to remove unwanted hydrocarbons. Because calciners operate at a higher temperature than dryers, they have the potential for volatilizing $^{210}$Pb and $^{210}$Po. Atmospheric releases from elemental phosphorus plants, which are mainly attributable to the operation of calciners, have been measured in the Netherlands and the United States [B31, U9, U10, U11, U12]. Estimated annual values are presented in Table 34. Discharges of $^{210}$Pb and $^{210}$Po are about two orders of magnitude higher than those of other solid radionuclides of the $^{238}$U decay series. Analyses of particles collected from the calciner off-gas streams at two plants in the United States showed that most of the $^{210}$Po activity was associated with submicron particles and that the dissolution rate of both $^{210}$Pb and $^{210}$Po in simulated lung fluid was very small [U10, U11].

225. Results of measurements at calciners at wet-process phosphoric acid plants are not yet available [U9]. Atmospheric discharges of natural radioisotopes at two wet-process phosphoric acid plants not equipped with calciners, however, have been published by EPA [P5] and summarized in the UNSCEAR 1982 Report [U1]. EPA has since defined the parameters of a reference wet-process phosphate fertilizer plant that are used to estimate the annual discharges of natural radionuclides into the atmosphere (Table 34). Those discharges result from the production processes of phosphate fertilizers and apparently do not include the discharges due to ore drying and grinding. Releases of $^{226}$Ra are estimated to be lower than those of $^{238}$U and $^{227}$Th because radium is chemically separated from uranium and thorium in the digestion.
of phosphate rock and goes predominantly to phosphogypsum.

226. Annual airborne discharges, normalized to the production or processing of one tonne of phosphate rock, are presented in Table 35. The normalized discharges apply mainly to the operation of phosphate industrial plants in the United States and may not be representative of the world-wide situation. Estimates of collective dose commitments corresponding to the normalized discharges have been calculated using the methods described in the section on coal-fired power plants and are also presented in Table 35.

227. Rough estimates of the collective dose commitments resulting from the world-wide operation of phosphate industrial facilities, using the experience of the United States as a guide, are presented in Table 36. The corresponding collective effective dose commitment is about 60 man Sv per year of practice.

228. Environmental concentrations of natural radionuclides and assessments of maximum individual doses have been published for a few plants. At a distance of about 2 km from an elemental phosphorus plant in the Netherlands, the activity concentrations of $^{210}$Pb and $^{210}$Po in surface air have been found to be about 0.1 and 0.4 mBq m$^{-3}$ above background, respectively [D7]. For the same plant, the estimated annual individual effective dose equivalent due to atmospheric discharges is about 40 $\mu$Sv [B31, S50]. In the United States, the atmospheric releases from the six elemental phosphorus plants in operation in 1983 were estimated to result in annual lung dose equivalents for nearby individuals ranging from 0.05 to 6 mSv [U9].

229. Discharges of radioactive materials into surface water have also been, occasionally reported. In the Netherlands, about 50 GBq of $^{210}$Pb and 30 GBq of $^{210}$Po were discharged from an elemental phosphorus plant into the Scheldt estuary [B31]. In that country, all phosphogypsum produced by fertilizer plants (2 million tonnes per year) is also disposed of into the Rhine 15 km from the North Sea [K19]; these annual discharges, which contain about 0.4 TBq of $^{238}$U, 2 TBq of $^{226}$Ra, 0.7 TBq of $^{210}$Pb and 2 TBq of $^{210}$Po, are estimated, in a preliminary assessment, to result in maximum individual effective dose equivalents of 150 $\mu$Sv per year and annual collective effective dose equivalents to the Dutch population of 170 man Sv from ingestion of seafood; the main contributor to the dose is $^{210}$Po [K19]. In France, over 3 million tonnes of phosphogypsum was dumped into the Seine estuary [P18] but the corresponding radiation exposures have not been estimated.

230. In other countries, such as India and the United States, phosphogypsum is retained in sludge ponds or stockpiles near phosphate fertilizer plants; occasional drainage or seepage of radioactive materials into nearby surface water has been reported [A14, P17].

231. Limited information is available on occupational exposures attributable to processing and transport of rock phosphates. In Dutch wet-process phosphoric acid plants, higher than average external radiation doses in air, ranging from 2 to 100 $\mu$Gy h$^{-1}$, have been detected in the vicinity of some piping and vessels near gypsum filters [L15]. Analysis of scaling samples revealed activity mass concentrations of $^{228}$Ra of up to 0.4 MBq kg$^{-1}$; the potential radiation doses, attributable to external exposure and to inhalation of $^{228}$Ra during periodic cleaning, have been estimated in eight phosphoric acid plants to be less than 2.2 mSv per year [L15]. In the Federal Republic of Germany, Pfister and Pauly [P15] estimated individual and collective external exposures for the small group of persons who handle rock phosphates as part of their job; their results correspond, in terms of effective dose equivalent, to individual exposures of 200 $\mu$Sv a$^{-1}$ and collective exposures of 0.5 man Sv a$^{-1}$. Extrapolating to the world's population by scaling over the phosphate fertilizer consumption results in an annual collective effective dose equivalent of approximately 20 man Sv. Exposures attributable to inhalation of phosphate rock, however, have not been taken into account.

2. Use of phosphate fertilizers

232. The world consumption of phosphate fertilizers was about 30 million tonnes of $\text{P}_2\text{O}_5$ in 1982 [F9]. The application rate of fertilizers depends on, among other things, the type of soil and the type of crop. The average consumption of phosphate fertilizer per unit area of agricultural land varied in 1982 from 3.6 kg $\text{P}_2\text{O}_5$ per hectare in the developing countries to 10.9 kg $\text{P}_2\text{O}_5$ per hectare in the developed countries; the world average being 6.7 kg $\text{P}_2\text{O}_5$ per hectare [F9].

233. Activity mass concentrations of natural radionuclides in phosphate fertilizers have been reviewed in the UNSCEAR 1982 Report [U1]. For a given radionuclide and type of fertilizer, the activity mass concentrations vary markedly from one country to another, depending on the origin of the components. General features are that the activity mass concentrations of $^{40}$K and of $^{232}$Th and its decay products are always low and that the activity mass concentrations of the radionuclides of the $^{238}$U decay series are 5-50 times higher than in normal soil. The degree of radioactive equilibrium between $^{238}$U and its decay products in a given type of fertilizer depends essentially on the relative contribution of phosphoric acid, since phosphoric acid usually has a very low $^{228}$Ra concentration. For the purposes of this Annex, it is assumed that $^{228}$Th and $^{232}$U are in radioactive equilibrium with $^{238}$U and that $^{210}$Pb and $^{210}$Po are in radioactive equilibrium with $^{228}$Ra. Table 37 presents average activity mass concentrations of $^{238}$U and $^{228}$Ra in phosphate fertilizer from four countries [D8, M21, P15, U13]; the levels range from 1.700 to 9.200 Bq per kg $\text{P}_2\text{O}_5$ for $^{238}$U and from 480 to 1.700 Bq per kg $\text{P}_2\text{O}_5$ for $^{228}$Ra. Typical values are 4,000 and 1,000 Bq per kg $\text{P}_2\text{O}_5$ for $^{238}$U and $^{228}$Ra, respectively.

234. Since the activity mass concentrations of $^{238}$U and $^{228}$Ra in phosphate fertilizers are several times higher than in normal soil, they constitute an additional
source of radiation exposure for workers and members of the public.

235. The annual application of phosphate fertilizers can be calculated to represent less than 1% of the normal soil content of $^{238}\text{U}$ [M21, P15]. Assuming an accumulation of $^{226}\text{Ra}$ in the soil during the past 80 years, Pfister and Pauly [P15] have estimated that the mean additional absorbed dose in air above fertilized fields is about 0.8 mGy h$^{-1}$, a small fraction of the normal natural background from terrestrial sources of 50 mGy h$^{-1}$. Small additional doses also occur from ingestion of foodstuffs grown on fertilized agricultural areas. Drichko et al. [D8] have shown that although the transfer coefficient of $^{226}\text{Ra}$ from soil to plants is practically the same whether $^{228}\text{Ra}$ is included in fertilizer or soil structures, the activity mass concentrations of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in the aerial parts of plants seem to be independent of the corresponding activity mass concentrations in soil; this implies the predominance of the air-to-vegetation transfer for such radionuclides.

236. The collective dose commitments incurred after application of phosphate fertilizers have been roughly evaluated by comparison with the activity mass concentrations of natural radionuclides in soil and the corresponding dose rates. Table 38 shows the results obtained under the following assumptions: the availability to plants of natural radionuclides is the same whether in fertilizer or in the normal constituents of the soil; the ploughed layer of soil is 0.3 m deep; the deposited activity becomes unavailable to the vegetation with a mean life of 100 years for the long-lived natural radionuclides; the fraction of time spent by the populations exposed on or near fertilized fields is 1%; and one tonne of phosphate rock produces, on average, 0.3 tonne of $\text{P}_2\text{O}_5$ in phosphate fertilizer. It can be inferred from Table 38 that the collective effective dose equivalent commitment per unit of phosphate rock is approximately 8 $10^{-2}$ man Sv t$^{-1}$. Taking the world-wide production of phosphate rock to be 120 million tonnes, the collective effective dose equivalent commitment resulting from the world-wide use of phosphate fertilizers during one year is roughly estimated to be 10,000 man Sv.

237. Occupational exposures arising from external irradiation during transport, storage and application of phosphate fertilizer were assessed by Pfister and Pauly [P15] and reviewed in the UNSCEAR 1982 Report [U1]. Mean additional absorbed dose rates in air ranging from 20 to 200 nGy h$^{-1}$ were measured for various transport and loading operations, with peak values of 800 nGy h$^{-1}$ [P15]. From the number of workers involved or the time spent in the various operations considered, a total annual collective effective dose equivalent of 1.3 man Sv can be derived [P15]. A rough estimate of about 50 man Sv for the collective effective dose arising from occupational exposure to external irradiation resulting from world-wide handling of phosphate fertilizers can be inferred from the results of Pfister and Pauly, using the consumption of phosphate fertilizers as a reference scale.

3. Use of by-products and wastes

238. The main by-products resulting from phosphate industrial activities are phosphogypsum in wet-process fertilizer plants and calcium silicate slag in thermal process plants. Significant radiation exposures may occur if such by-products are used in the building industry. Another source of exposure stems from the reclamation of land on which phosphate mining has been completed and houses have been built. Since new information on radiation exposures has not been made available to the Committee in the past few years, the following is, to a large extent, a summary of the corresponding section in the UNSCEAR 1982 Report [U1].

239. Large quantities of phosphogypsum (about 100 million tonnes per year) are produced in wet-process phosphoric acid plants. The activity mass concentrations of $^{226}\text{Ra}$ in phosphogypsum, which depends on the origin of the phosphate ore processed, is typically about 900 Bq kg$^{-1}$ [U1, U2]. Most of the phosphogypsum is considered waste and is either stored in ponds or stacks or discharged into the aquatic environment.

240. In the United States, a small amount of phosphogypsum (about 0.6 million tonnes per year) is used in agriculture to improve water movement in saline-alkaline soil and as a substitute for lime or limestone in alkaline soil [L16, P18].

241. Phosphogypsum is also used as a substitute for natural gypsum in the manufacture of cement, wallboard and plaster. Japan phased out its production of natural gypsum in 1976 and is currently recycling about 3 million tonnes of phosphogypsum per year [P18]. In Japan, approximately one half of the by-product gypsum is used as cement set-retardant; the other half is used to make wallboard and plaster. In Western Europe, about 10% of the phosphogypsum production, that is 2.5 million tonnes, was used in 1981 as by-product [P18]. Phosphogypsum is commonly used in construction blocks for floors and walls and in partition blocks for interiors in North Africa, the Mediterranean countries and the Middle East. It will be assumed in this Annex that 5 million tonnes of phosphogypsum, representing 5% of the world-wide annual production, is used annually as building material in dwellings.

242. O’ Riordan et al. [O8] estimated the additional doses that would be received by the occupants of a residential building in which 4.2 tonnes of by-product gypsum would have replaced the established materials; the additional absorbed dose rate in air was calculated to be 0.07 $\mu$Gy h$^{-1}$; the additional radon concentration, using an air exchange rate of 1 h$^{-1}$, was estimated to be about 10 Bq m$^{-3}$, resulting in an annual effective dose equivalent of 0.6 mSv. If it is assumed that 5% of the by-product gypsum is used as building material in dwellings, on average four persons live in the dwellings and the mean life of the dwellings is 50 years, the collective effective dose equivalent commitments resulting from one year of world-wide use of phosphogypsum in the building industry are
estimated to be $10^4$ man Sv from external irradiation and $2 \times 10^3$ man Sv from internal irradiation. These estimates are highly uncertain and need to be confirmed by measurements in dwellings that have been constructed using known amounts of phosphogypsum.

243. Calcium silicate slag may be used as a component for concrete. Measured activity concentrations in slag samples range from 1.300 to 2,200 Bq kg$^{-1}$ of $^{226}$Ra [B11, M16]. Results from an indoor survey indicate that the gamma absorbed dose rate in air can be as high as 0.3 $\mu$Gy h$^{-1}$ above background in dwellings constructed with 43% by weight slag in concrete slabs [B11]. In a similar survey carried out in Canada, absorbed dose rates of up to 0.2 $\mu$Gy h$^{-1}$ were obtained [M16].

244. Residents in structures built on reclaimed phosphate land in Florida, United States may also be exposed to significant radiation doses. About 600 km$^2$ of Florida land has been mined for phosphate rock in the past 80 years. About 200 km$^2$ of the land has been reclaimed to various degrees. The equilibrium equivalent radon concentrations in houses built on reclaimed land were found to range from 4 to 500 Bq m$^{-3}$ with a weighted average of about 40 Bq m$^{-3}$, which is estimated to be 26 Bq m$^{-3}$ above normal concentrations in houses in that area [G21]. This average additional concentration results in an annual effective dose equivalent of approximately 1.6 mSv. An experimental investigation and remedial action programme was carried out involving 10 houses and a mobile home on land that had phosphate mineralization or had been affected by phosphate mining activities and for which the annual average radon equilibrium equivalent concentration was believed to be in excess of 100 Bq m$^{-3}$ [S51].

4. Summary

245. Table 39 summarizes the radiation exposures attributable to the use of phosphate rock, expressed in terms of collective effective dose equivalent commitment resulting from one year of practice. The total collective effective dose equivalent commitment is estimated to be 300,000 man Sv, essentially from the use of by-product gypsum in dwellings.

D. NON-URANIUM ORE MINING AND PROCESSING

246. Because of the presence of radon in confined spaces, metal and non-metal mining results in occupational exposures. Ventilation of mine air, in turn, results in environmental discharges of radon and exposures of members of the public. Ore processing gives rise to additional environmental discharges of natural radionuclides. Occupational exposures and exposures of members of the public are discussed below.

1. Occupational exposures from non-uranium mining

247. Investigations of radon and radon decay product concentrations in non-uranium mines have been conducted since the 1960s. This section is devoted to the discussion of occupational exposures in non-uranium mines other than coal mines, which are considered in another part of this Annex.

248. Measured concentrations of radon and radon decay products are presented in Table 40. The potential alpha energy concentrations of radon decay products range from 0.02 to more than 300 $\mu$J m$^{-3}$, and there is no simple relationship between radon decay product concentration and type of mine. Mines extracting from sedimentary deposits usually have low radon decay product concentrations but this is not always true. Mines associated with igneous intrusions imply high uranium content and therefore high radon decay product concentrations, although ventilation can obscure this relationship. Geological and hydrogeological characteristics may also be of importance. Convective movements of air and water resulting from pressure differences between the mine and its environment explain large radon entry rates into the mine [Z2]. Detailed studies of the characteristics of non-uranium mines in countries of the European Economic Community show that the primary indicator of high radiation exposures is a radon concentration in exhaust air in excess of 400 Bq m$^{-3}$ [Z2].

249. Occupational exposures attributable to radiation in non-uranium mines have decreased with time [D6, S34, S35], as monitoring has led to action being taken to relieve high exposure situations. Personal dosimetry is now being used in Poland non-uranium mines [C15] and is envisaged in Italy [S37] and the United Kingdom [M18].

250. The degree of equilibrium between radon decay products and radon in non-uranium mines has been investigated in several countries. Measurements in an Italian lead mine at a location representative of the radiological characteristics of the mine show an average value of 0.70 with a range of 0.50-0.85 [S37]. In 15 mines in the Federal Republic of Germany, the equilibrium factor was found to vary from 0.28 to 0.82 with a rather flat distribution [K15]. In Norway, the mean value of 210 measurements of the equilibrium factor in different mines was 0.46, with a minimum of 0.08 and a maximum of 0.93 [S38]. A typical value of the equilibrium factor in non-uranium mines seems to be 0.6 with large variations around this figure.

251. Stranden and Berteg [B20, S38] have established empirical relationships between the equilibrium factor $F$ and the activity concentrations $\chi$ of the individual radon decay products $^{218}$Po, $^{214}$Pb and $^{214}$Bi normalized to radon that seem to apply to most mining conditions when the value of $F$ is in the range of 0.1-0.7:

$$\chi^{(218}\text{Po}) / \chi^{(222}\text{Rn}) = 1.1 F^{0.55}$$
$$\chi^{(214}\text{Pb}) / \chi^{(222}\text{Rn}) = 1.0 F^{0.94}$$
$$\chi^{(214}\text{Bi}) / \chi^{(222}\text{Rn}) = 0.77 F^{1.12}$$

252. The unattached fraction $f_a$ of the potential alpha energy of radon decay products in non-uranium mines was also studied by Berteg and Stranden [B20, S38]; in most working conditions, the value of $f_a$ is usually found to be smaller than 0.05. The main factor
influencing the unattached fraction seems to be the aerosol particle concentration: high values of the unattached fraction are observed with low aerosol concentrations [G23, S38]. Suggested empirical relationships between the value of $f_p$ and that of the unattached fraction of individual radon decay products are as follows [S38]:

$$f^{(210)Po} = 2.6 f_p^{(-1)}$$
$$f^{(214)Pb} = 0.77 f_p^{(-1)}$$
$$f^{(214)Bi} = 0.27 f_p^{(-1)}$$

253. Radiation measurements other than those of radon and their decay products are limited. A study of the complete spectrum of radiation exposures was carried out in eight British non-uranium mines [D5]. Table 41 presents the reported concentrations and an estimate of the resulting effective dose equivalents. In mines associated with igneous intrusions, the calculated doses are almost entirely due to the inhalation of radon decay products. In sedimentary mines, the contribution to the normal dose from the inhalation of radon decay products is always greater than 50%, even for low concentrations of radon daughters. Similar results have been observed for mines in the Federal Republic of Germany [K15]. The effective dose equivalent from inhaled thoron decay products, however, may be higher than that from inhaled radon decay products if the ratio of the activity mass concentration of $^{228}$Th to that of $^{230}$U in the mined rock is substantially greater than one, which is sometimes the case [S39, S40].

254. On the basis of the results presented in Table 40, annual effective dose equivalents received by non-uranium miners in the course of their work are estimated to range from 0.1 mSv to about 0.2 Sv, most of the values being between 1 and 50 mSv. If the arithmetic mean of the annual effective dose equivalent is tentatively assumed to be 10 mSv and the fraction of non-uranium miners in the world population is taken to be $5 \times 10^{-4}$ [U14], a very rough estimate of the annual collective effective dose equivalent would be $2 \times 10^6$ man Sv.

2. Occupational exposures from factory processes

255. Industrial processes are carried out for many purposes that use the materials extracted by non-uranium mining. In these factory processes, there are exposures to natural radioactivity but in most cases data on measured activities or estimates of doses have not been reported in the literature. Examples given in the following sections are the industrial use of zircon sand and the processing of ilmenite; another example not discussed separately in this Annex is the use of thorium in coating optical lenses. Information in this field is extremely limited and no reliable assessment of the collective exposures can be made.

(a) Processing of raw ilmenite

256. The activity concentrations of natural radionuclides in samples collected at an Australian titanium dioxide plant have been reported [C13]. In that plant, the white pigment titanium dioxide is produced from raw ilmenite (a dark-coloured, fine-grained beach sand) containing monazite as an impurity (0.14% by mass). The activity concentrations in ilmenite feed are about 400 Bq kg$^{-1}$ of $^{228}$Ra and $^{228}$Th and 75 Bq kg$^{-1}$ of $^{238}$Ra. These radionuclides have been followed throughout the pigment production process. It was established that in the presence of high sulphate ion concentrations in both the plant feed liquor and the effluent, thorium remains in solution whereas radium, which forms an insoluble sulphate compound, tends to co-precipitate and is found in solid residues throughout the process. Very high activity concentrations of $4 \times 10^5$ Bq kg$^{-1}$ of $^{228}$Ra and $1.5 \times 10^5$ Bq kg$^{-1}$ of $^{228}$Th were measured in a sample of scale scraped from a heat exchanger unit. The mass balance study of the activity entering and leaving the plant shows, however, that essentially all of the activity entering the plant (14 MBq d$^{-1}$ of $^{228}$Ra and 67 MBq d$^{-1}$ of $^{228}$Th) is found in the acidic liquid effluent stream, which is pumped to disposal lagoons and is allowed to neutralize and drain away into an estuary. Effluent concentrations are about 6 Bq l$^{-1}$ of $^{228}$Ra and $^{228}$Th and 1.4 Bq l$^{-1}$ of $^{228}$Ra, whereas in the estuary they are approximately 0.01 Bq l$^{-1}$ for $^{228}$Ra, 0.02 Bq l$^{-1}$ for $^{228}$Th and 0.1 Bq l$^{-1}$ for $^{228}$Ra. In contrast to the situation found in the plant and the effluent stream, radium is present in the estuary in a soluble form and thorium is not, leading to a possible accumulation of $^{232}$Th and of its decay products in the estuary. The radiation doses attributable to the operation of the plant have not been assessed.

(b) Industrial use of zircon sand

257. Zircon sand occurs in river and beach placers, usually with monazite. About 700,000 tonnes of zirconium minerals are produced each year, mainly in Australia and South Africa [B37]. The activity mass concentrations of $^{238}$U and $^{232}$Th in zircon sand are in general well above 500 Bq kg$^{-1}$ [B36, D13, N18]. Zircon sand is used in the zirconium extraction industry, foundry sands, refractories and ceramics. Use of the sand by foundries and the zirconium industry has resulted in the accumulation of large volumes of low-level radioactive waste.

258. Little information is available on the radiation protection aspects of the use of zircon sand [B36, N18]. It seems clear, however, that inhalation of radon decay products is not a problem because the exhalation rate of radon from the zircon sand is low. Doses from external irradiation may be significant only in storage areas. Absorbed dose rates in air of about 700 nGy h$^{-1}$ have been measured at a distance of about 1 m from a stack of bags of zircon sand [B36]. In an Italian factory producing electro-fused refractory blocks, the absorbed dose rates in air were observed to be near background in all representative work places but reached about 500 nGy h$^{-1}$ on top of the sand storage tanks [N18].

259. The inhalation pathway, also assessed in the Italian factory, seems to be more important. The airborne concentrations of natural radionuclides near areas of high temperature processes were estimated
to be about 200 mBq m$^{-3}$ of $^{210}$Po (typical background: 0.04 mBq m$^{-3}$) and 20 mBq m$^{-3}$ for very long-lived natural radionuclides (typical background: 0.001 mBq m$^{-3}$). The particle size distribution was bimodal with a mode at approximately 0.3 μm and another between 5 and 10 μm. The resulting annual effective dose equivalent, assuming constant occupancy in the most highly contaminated areas, would be of the order of 5 mSv [N18].

260. No information on environmental discharges was made available to the Committee. Aquatic discharges would in most cases lead only to insignificant doses, as zircon compounds are extremely insoluble in water.

3. Exposures of members of the public

261. All industrial operations involving ore mining and processing release natural radionuclides into the environment. The results of radiation surveys conducted in several industrial facilities are given below.

(a) Zinc mining and processing

262. Zinc is usually found in nature as a sulphide ore called sphalerite. The ore, which usually contains impurities of lead, cadmium and traces of other elements, are processed at the mill to form concentrates typically containing 62% zinc and 32% sulphur. These concentrates are processed at the smelter to recover zinc metal [U8].

263. Atmospheric emissions of naturally occurring radionuclides were measured from an underground mine and mill [A7] and estimated from a reference zinc smelter [U8]: the results are shown in Table 42. The releases from the smelter cannot be compared to those of the mine and mill, as they are different facilities and the capacity of the mine and mill was not indicated. It is clear, however, that the only significant emission is that of radon from the mine (8 TBq per year). A radiation survey was conducted in that particular zinc mine because a relatively high radon concentration of 2,600 Bq m$^{-3}$ had been measured at the mine portal exhaust. Concentrations of $^{238}$U and $^{232}$Th, in the ore as well as in the concentrate, were found to be significantly below the corresponding average concentrations in soil; the relatively high radon concentrations observed in the mine were attributed to the large quantity of water that flows into the mine. The collective effective dose equivalent commitment resulting from inhalation of radon decay products released in one year was estimated to be about 0.1 man Sv. The very small releases of other natural radionuclides due to low activity mass concentrations and to efficient particle control do not result in significant collective effective dose equivalent commitments.

(b) Fireclay mine and refractory plant

264. Clay mining and manufacturing is a large industry. Clay may be used to manufacture bricks and refractory materials. A clay mine and refractory plant was surveyed in the United States to estimate the quantities of naturally occurring radionuclides emitted into the atmosphere during mining and manufacturing processes [A6]. Ore samples collected from the mine contained average $^{238}$U and $^{232}$Th concentrations of about 50 and 70 Bq kg$^{-1}$, respectively. Equilibrium equivalent concentrations of radon of approximately 1 kBq m$^{-3}$ were measured in the clay mine at the working face; the corresponding annual emission of radon is about 1 TBq. Particulate releases consist mainly of $^{210}$Po, which is partially vaporized at the kiln's operating temperature of 1,100° C. Annual atmospheric emissions are summarized in Table 43. Estimated collective effective dose equivalent commitments resulting from one year of operation of the fireclay mine and refractory plant are 0.02 man Sv from inhalation of radon decay products and less than 0.01 man Sv from inhalation and ingestion of $^{210}$Po.

(c) Aluminium ore processing

265. Bauxite is the principal aluminium ore found in nature. The ore is processed at mines to produce alumina (Al₂O₃), the basic feed in the aluminium reduction process. Aluminium metal is produced by the reduction of alumina in a molten bath of cryolite [U8].

266. Radionuclide measurements at both an alumina plant and an aluminium reduction plant have been carried out in the United States [A9, U8]. The $^{232}$Th activities per unit mass measured in the process samples of the alumina plant are listed in Table 44. The levels in bauxite ore, red mud and brown mud are about 10 times higher than the average activities per unit mass in normal soil, whereas the radioactive content of alumina kiln feed and alumina product is less than that of normal soil.

267. Table 45 shows the estimated annual atmospheric emissions from the surveyed alumina plant and aluminium reduction plant. Again, radon emissions are much higher than those of other natural radionuclides. Among the particulates, $^{210}$Pb and $^{210}$Po are the most important contributors to the environmental releases, as the high temperatures of the kilns and of the reduction cells (above 950° C) cause these radionuclides to be volatilized [A9, U8].

268. The corresponding collective effective dose equivalent commitments are estimated to be about 0.1 man Sv; they are almost entirely due to the releases of $^{210}$Po and $^{210}$Pb.

(d) Copper ore mining and processing

269. Copper ores are milled to produce a concentrate containing copper, sulphur, iron and some insoluble material (primarily silica and aluminium). This concentrate is the basic feed to the copper smelter that eventually produces the refined copper product. Prior to smelting, part or all of the concentrates may receive a partial roast to eliminate some of the sulphur and other impurities [U8].

270. Radionuclide measurement studies at an underground copper mine and mill, an open pit mine and mill and two copper smelters have been carried out in
the United States [A9, U8]. The activity mass concentrations of $^{238}$U and $^{222}$Th in process samples from the mines and mills (Table 46) were found to be at or slightly above the corresponding levels in normal soil. Most of the $^{232}$Th content, however, seems to be removed from the ore in the milling process.

271. Estimated annual atmospheric releases are presented in Table 47. Emissions from the copper smelter cannot be directly compared with those from the mine and from the mill, the capacity of which were not indicated. Calculated collective effective dose equivalent commitments from one year of releases are about 1 man Sv, with nearly equal contributions from thorium isotopes and $^{210}$Pb-$^{210}$Po.

(e) Lead ore processing

272. Galena (PbS), frequently containing cerussite (PbCO$_3$) and anglesite (PbSO$_4$), is the principal lead-bearing ore found in nature. Galena contains small amounts of copper, iron, zinc and other trace elements. Lead smelting involves three distinct processes: sintering, to convert the ore from a sulphide to an oxide or sulphate form and prepare the feed materials for furnacing; furnacing, to reduce the oxide feed to lead metal; and dressing, to reduce the copper content of the lead bullion from the furnace. Off-gases from the sintering machine and the blast furnace are the most significant sources of particulate emissions from the lead smelting process [U8].

273. As the activities of $^{214}$U and $^{212}$Th per unit mass of lead ore are similar to the corresponding values in normal soil, the atmospheric emissions of natural radionuclides estimated for a reference plant processing annually 0.22 million tonnes of lead [U9] are fairly small (about 1 GBq of $^{210}$Pb and of $^{210}$Po). The resulting collective effective dose equivalent commitments per year of release from the reference plant are less than 0.1 man Sv.

(f) Other metal and non-metal mining

274. Annual radon releases into the atmosphere have been reported for a few other United States mines: 30 GBq for a limestone mine, 70 GBq for a fluor spar mine and 2,600 GBq for an iron mine [S32]. The resulting collective effective dose equivalent commitments from inhalation of radon decay products are less than 0.05 man Sv.

IV. SUMMARY

275. Estimates of per caput annual effective dose equivalents from external irradiation due to cosmic rays and terrestrial sources and from internal irradiation due to the most important naturally occurring radionuclides are presented in Table 48, together with the subjectively estimated typical ranges of variation for all the sources taken into consideration. By far the most important source of exposure is indoor radon, which leads to a per caput effective dose equivalent of 1,100 $\mu$Sv and to a wide range of variation. The overall per caput annual effective dose equivalent is estimated to be about 2,400 $\mu$Sv, corresponding to an annual collective effective dose equivalent of approximately $10^5$ man Sv. The current estimate of the overall per caput annual effective dose equivalent is 400 $\mu$Sv higher than the estimate provided in the UNSCEAR 1982 Report. This is mainly the result of a re-evaluation of the mean indoor radon concentration. The wide distribution of individual effective dose equivalents is dominated by the variability of the indoor radon concentrations, which span several orders of magnitude.

276. Individual exposures resulting from industrial activities are generally small in comparison to the overall exposure from natural sources of radiation. Their importance can best be characterized in terms of collective effective dose equivalent commitments. Table 49 provides estimates of collective effective dose equivalent commitments arising from one year of practice for most of the activities considered. Because of the scarcity, or lack, of data for the industrial activities listed in the table, most of the estimates are very tentative. The total collective effective dose equivalent commitment arising from one year of practice is less than $5 \times 10^4$ man Sv, representing a per caput effective dose equivalent commitment of about $100 \mu$Sv.