1. Exposure Data

1.1 Environmental exposure

1.1.1 *Detonation of nuclear weapons*

(*a*) *Sources*

The most important source of radioactive fall-out from nuclear explosions is the large number of nuclear weapons tests carried out in various parts of the world since 1945. A total of 543 identified atmospheric tests have been conducted worldwide. The largest number of atmospheric tests was carried out in the 1950s and in 1961–62 by the former Soviet Union (USSR) and the United States of America (USA). These countries and the United Kingdom ended their atmospheric testing after signing a limited test ban treaty (banning atmospheric tests) in 1963, but France and China conducted additional tests — although fewer — until 1974 and 1980, respectively. A larger number of underground nuclear tests (over 1850) have been carried out, but these resulted in less human exposure than the atmospheric tests. The majority of the underground tests were conducted after 1963 (Bouville *et al.*, 2000; UNSCEAR, 2000).

Fall-out can occur from other nuclear explosions. About 100 underground nuclear explosions have been carried out for peaceful purposes, such as excavation, mining and cratering, in the USA and the former USSR. The estimated collective doses from these peaceful nuclear explosions are very low. A similarly low collective dose is estimated to have resulted from the burn-up and re-entry of satellite power sources, which are commonly fuelled with ^{238}Pu (UNSCEAR, 1993). Another source is the wartime use of atomic bombs over Hiroshima and Nagasaki, which primarily resulted in acute exposure to γ-rays and neutrons emitted directly from the bombs. The health consequences of acute radiation have been the subject of extensive and continuing epidemiological investigations (see e.g. IARC, 2000). The fall-out radiation in Hiroshima and Nagasaki has been less well characterized than direct exposure but is considered to have been small.

In atmospheric tests, particles containing radionuclides are released during the explosion and are carried into the stratosphere, giving rise to worldwide fall-out. Human exposure to external irradiation occurs when the radionuclides are deposited on the earth's surface, and internal exposures occur when the radionuclides are incorporated into the body through ingested foods. Although a well-contained underground nuclear explosion delivers extremely low doses, on some occasions venting or diffusion of gases or liquids has resulted in leakage of radioactive materials after underground tests, leading to regional dissemination of radioactive debris (UNSCEAR, 1993).

(*b*) *Global exposures*

Exposures to radiation from nuclear weapons tests have been a major concern of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), which assesses and updates data on exposure on a global basis. The basic quantity used by UNSCEAR to express the radiation doses imposed on the world's population is the 'collective dose commitment'. This is the integral over infinite time of the collective dose rates delivered to the world's population, i.e. including doses to be delivered in future time until complete decay or removal of the radionuclides from the environment. Calculated in this way, collective doses have little meaning with respect to the dose received by the first generations after radionuclide release, as the collective dose is dominated by doses received from a few long-lived isotopes, e.g. ^{14}C and ^{237}Np, which deliver only a very small dose to individuals. It follows that it might be more appropriate to truncate the calculation of collective dose when the dose rate becomes insignificant. Currently, fall-out accounts for only 0.2% of the average background dose in the United Kingdom (National Radiological Protection Board, 2000). Sources of internal irradiation are inhalation of air contaminated with radionuclides and ingestion of contaminated foodstuffs. Internal exposures, especially through ingestion, are the major component of the total effective doses committed by the tests on a global basis. UNSCEAR estimated that the total effective dose commitment from atmospheric nuclear testing to the world's population is 30×10^6 person–Sv, of which about 93% (28×10^6 person–Sv) is from internal exposure due to ingestion or inhalation of radionuclides (Table 1) (UNSCEAR, 1993). By far the most important component is ingested ^{14}C, which accounts for almost 26×10^6 person–Sv because of its very long half-life (5730 years) and environmental mobility. This radionuclide will deliver very small dose rates to the world population during thousands of years at the same rate as it does now; it is the accumulation of these very small dose rates over a long period to a large population (including the future population) that explains the large contribution of ^{14}C. ^{137}Cs, which is the most important component (about 1.2×10^6 person–Sv) of external doses from the nuclear tests, is the second most important component of the internal doses (about 0.7×10^6 person–Sv) committed by the nuclear tests. This is followed by ^{90}Sr, which contributes a little over 0.4×10^6 person–Sv (IARC, 2000). These estimates do not include the exposure from local fall-out, discussed below, of people who live near the test sites and thus may receive relatively high doses.

(*c*) *Local and regional exposures*

Nuclear tests are conducted at isolated sites when the meteorological conditions are favourable, but unexpected events, such as the shift in winds during the Bravo test at Bikini Atoll in the Marshall Islands (see below), can result in heavy radiation exposure from local fall-out. Local and regional exposure doses have been estimated for several major nuclear test sites, including that in Nevada (USA), in the Pacific

Table 1. Collective effective dose commitment from atmospheric nuclear tests to internal exposure of the world's population

Radionuclide	Half-life	Collective effective dose commitment (1000 person–Sv)		
		Ingestion	Inhalation	Total internal exposure
^{14}C	5730 years	25 800	2.6	25 800
^{137}Cs	30.1 years	677	1.1	678
^{90}Sr	28.8 years	406	29	435
^{3}H	12.3 years	176	13	189
^{131}I	8.02 days	154	6.3	160
^{144}Ce	285 days		122	122
^{106}Ru	374 days		82	82
^{239}Pu	24 100 years	1.8	56	58
^{241}Am	432 years	8.7	44	53
^{240}Pu	6537 years	1.3	38	39
^{55}Fe	2.73 years	26	0.06	26
^{241}Pu	14.4 years	0.01	17	17
^{89}Sr	50.5 days	4.5	6.0	11
^{91}Y	58.5 days		8.9	8.9
^{95}Zr	64.0 days		6.1	6
^{95}Nb	35.0 days		2.6	2.6
^{103}Ru	39.3 days		1.8	1.8
^{140}Ba	12.8 days	0.81	0.66	1.5
^{141}Ce	32.5 days		1.4	1.4
^{54}Mn	312 days		0.4	0.4
^{125}Sb	2.76 years		0.2	0.2
Total (rounded)		27 300	440	27 700

From UNSCEAR (1993)

(Marshall Islands), Semipalatinsk and Novaya Zemlya in the former USSR, Lop Nor in China, Mururoa and Fangataufa (French test sites) and Australian test sites (used by the United Kingdom).

The doses to most organs and tissues from ingestion and inhalation after local fall-out are substantially lower than those from external exposure, with the important exception of doses to the thyroid, for which internal exposure to radioactive iodines predominates. The dose that the thyroid receives from internal exposure is often greater than that from external exposure or the doses that any other organ receives from internal exposure (Whicker *et al*., 1996; UNSCEAR, 2000).

(i) *Nevada test site, USA*

Between 1951 and 1962, at least 105 atmospheric tests were conducted at the Nevada test site, and 14 other tests at depths where containment was not expected. These resulted in the atmospheric release of ^{131}I, ^{137}Cs and other radionuclides. Extensive dose reconstruction has been undertaken for the population living in the vicinity of the test site during the period of atmospheric testing. In particular, the Off-site Radiation Exposure Review Project of the Department of Energy in the USA collected information on fall-out in off-site areas and provides dosimetric data by region, community, locale, age and occupation. The doses from external irradiation have been calculated by Monte Carlo techniques for each event for the residents of each town in the vicinity of the site and for each county in the affected region. The doses from internal exposure have been estimated by pathway models for combinations of location and event for various radionuclides, age groups and organs. The data from these projects, supplemented by additional work, were used in epidemiological studies on thyroid cancer and leukaemia in Utah (Stevens *et al.*, 1990; Kerber *et al.*, 1993; Simon *et al.*, 1995; Till *et al.*, 1995). In the study on thyroid cancer, special efforts were made to estimate individual doses to the thyroid from data on diet and lifestyle obtained from a survey.

Anspaugh *et al.* (1990) estimated the doses to the thyroid for an infant living in St George, Utah, when the event HARRY occurred on 19 May 1953, which resulted in fall-out over St George. This was one of the most heavily contaminated areas, and the test accounted for most of the doses to the thyroid in that area. Table 2 shows that previous estimates are comparable to that of Anspaugh *et al.* (1990).

The National Cancer Institute (1997) conducted a study to estimate the doses received by the thyroid for people living across the contiguous USA as a result of fall-out of ^{131}I from the Nevada test site. Table 3 presents the per-capita thyroid doses (summed over all Nevada test site events) estimated in the study. The results indicate that deposition of ^{131}I occurred at one time or another in every county of the contiguous USA between 1951 and 1958. The estimated collective thyroid dose was about 4×10^6 person–Gy, with a per-capita dose of about 20 mGy.

(ii) *Pacific test site, Marshall Islands*

The USA conducted at least 105 tests in the Pacific region between 1946 and 1962. In terms of radiation exposures, the tests conducted at Bikini atoll in the Marshall Islands were the most important. Of special importance is the Bravo thermonuclear test conducted in March 1954. Following this test, an unpredicted shift in winds resulted in exposure to radioactive fall-out of some 250 inhabitants of the Marshall Islands, 28 American servicemen on atolls to the east and 23 Japanese fishermen on their fishing vessel (Conard *et al.*, 1980; Bouville *et al.*, 2000).

The health effects on the Marshallese who resided on three atolls, Rongelap, Ailinginae and Utirik, at the time of detonation have been studied. These atolls were 100–300 miles from the detonation site. Exposure was largely from fall-out deposited

Table 2. Doses to the thyroid for an infant living in St George, Utah, USA, at the time of the event HARRY on 19 May 1953

Thyroid dose (Gy) (central estimate and/or range of uncertainty)	Reference
0.68	Mays (1963)
1–7	Reiss (1963)
0.84	Pendleton *et al.* (1963)
1.2–4.4	Knapp (1963)
0.78 (0.2–1.6)	Tamplin & Fisher (1967)
0.68	Perez & Robinson (1967)
0.66 (0.2–1.9)	Ng *et al.* (1990)
0.5 (0.2–1.4)	Anspaugh *et al.* (1990)

From Anspaugh *et al*. (1990)

Table 3. Estimated collective doses to the thyroid for the population in the USA due to fall-out from the Nevada atmospheric nuclear bomb tests

Series	Dates	Collective thyroid dose (person-Gy)
Ranger	January–February 1951	1.6×10^3
Buster-Jangle	October–November 1951	7.4×10^4
Tumbler-Snapper	April–June 1952	1.1×10^6
Upshot-Knothole	March–June 1953	8.9×10^5
Teapot	February–May 1955	4.1×10^5
Plumbbob	May–October 1957	1.2×10^6
Hardtack II	September–October 1958	1.6
All		3.7×10^6

From National Cancer Institute (1997)

on the skin and internal deposition of radionuclides from ingestion of contaminated food and water and involved a mixture of radionuclides (^{131}I, ^{132}I, ^{133}I, ^{134}I and ^{135}I), tellurium isotopes and γ-rays (Lessard *et al.*, 1985; UNSCEAR, 2000). The most extensive evaluation of doses to the thyroid was carried out by Lessard *et al*. (1985), on the basis of measurements of ^{131}I in a pooled urine sample collected on the 17th day after detonation of the Bravo bomb. The estimated doses, largely due to internal exposure (roughly 80–90%), were highly dependent on age (Table 4), infants receiving the highest doses. Although short-lived radioiodines (^{131}I, ^{133}I, ^{135}I) were not

Table 4. Estimated doses to the thyroid from internal and external exposure after the Bravo test in the Marshall Islands

Atoll	Age	Estimated dose (Gy)		
		Internal	External	Total
Rongelap	Adult	10	1.9	12
	9 years	20	1.9	22
	1 year	50	1.9	52
	Newborn	2.5	1.9	4.4
	In utero	6.8	1.9	8.7
Ailinginae	Adult	2.8	1.1	4.0
	9 years	5.4	1.1	6.6
	1 year	13.0	1.1	14.0
	In utero	4.9	1.1	6.1
Utirik	Adult	1.5	0.11	1.6
	9 years	3.0	0.11	3.1
	1 year	6.7	0.11	6.8
	Newborn	0.48	0.11	0.59
	In utero, 3rd trimester	0.98	0.11	1.1
	In utero, 2nd trimester	2.6	0.11	2.7

From Lessard *et al*. (1985)

measured, half of the dose was considered to be due to the intake of ^{133}I, while ^{131}I contributed 10–15% of the doses at Rongelap and Ailinginae and about 20% at Utirik (Lessard *et al*., 1985).

(iii) *Semipalatinsk test site, Kazakhstan*

At the Semipalatinsk test site in the Republic of Kazakhstan, in the former USSR, 116 uncontained nuclear and thermonuclear explosions were set off, starting in 1949. Most of the regional radioactive contamination, outside the bounds of the test site itself, was due to tests conducted between 1949 and 1956, contributing more than 95% of the expected collective doses of the non-occupationally exposed population living close to the test site (UNSCEAR, 1993; Dubasov *et al.*, 1994; UNSCEAR, 2000).

Several groups of investigators are reconstructing the doses from external and internal exposures of the population living in the vicinity of the test site. About 10 000 people living in the settlements bordering the site are reported to have been exposed to some extent (UNSCEAR, 1993, 2000). The total collective dose attributable to the tests during 1949–62 is estimated to be 2600 person–Sv from external irradiation and 2000 person-Sv from internal exposure due to ingestion of radionuclides. For internal exposure, the collective doses are estimated to be about 10 000 person–Sv to the thyroid and about 2000 person–Sv to bone marrow (UNSCEAR, 1993; Bouville *et al.*, 2000).

(iv) *Novaya Zemlya, Russian Federation*

More than 90 atmospheric nuclear tests were conducted on the Novaya Zemlya islands, Russian Federation, and these account for about half of the total energy yield of all nuclear tests carried out worldwide. Nevertheless, the local doses to off-site residents are considered to be relatively low, as most of the atmospheric explosions were conducted at high altitudes, most of the vented underground tests resulted in on-site contamination only, and the test site is large and isolated. Little information is available on local and regional doses, and then only on on-site contamination and doses received by reindeer herders and people who consume reindeer meat (UNSCEAR, 2000).

(v) *Lop Nor, China*

China conducted 22 atmospheric tests between 1964 and 1980 at the Lop Nor test site. The estimated effective doses from external exposure of populations 400–800 km downwind from the test site ranged from 0.006 to 0.11 mSv, with a mean of 0.044 mSv (UNSCEAR, 2000). The doses to the thyroid from internal exposure to ^{131}I for adults ranged from 0.059 to 2.5 mGy. The average dose to the thyroid received by the Chinese population from these tests is estimated to be about 0.14 mGy (Bouville *et al.*, 2000).

(vi) *Mururoa and Fangataufa, French Polynesia*

France conducted 46 atmospheric tests on the uninhabited atolls of Mururoa and Fangataufa, French Polynesia, between 1966 and 1974. About 5000 people lived within a 1000-km radius of the planned ground zero in Mururoa. After 1975, all tests were conducted underground. Doses have been reconstructed for the 110 000 inhabitants of Tahiti, located 1000 km from Mururoa and Fangataufa, and for the 140 inhabitants of the Tureia atoll. The doses to the thyroid from internal exposure to ^{131}I of infants in Tahiti during the atmospheric testing were estimated to be 0.12–6.8 mGy, the highest occurring in 1974 (UNSCEAR, 1977; Bouville *et al.*, 2000; UNSCEAR, 2000).

(vii) *Emu, Maralinga and Montebello, Australia*

The United Kingdom conducted 12 atmospheric tests in Australia between 1952 and 1957. The doses from internal exposure have been assessed on the basis of the estimated ingestion of fall-out radionuclides in food and drinking-water and inhalation of fall-out radionuclides in air. For the entire Australian population, the average effective dose from these tests was estimated to be 0.07 mSv and the collective effective dose equivalent to be 700 person–Sv (UNSCEAR, 2000).

1.1.2 *Accidents at nuclear installations*

(*a*) *Chernobyl*

During an engineering test of one of the four reactors at the Chernobyl nuclear power plant in the Ukraine on 26 April 1986, the safety systems had been switched off, and unstable operation of the reactor allowed an uncontrollable power surge to occur, leading to successive steam explosions and resulting in destruction of the reactor. Within days or weeks of this accident, 28 power-plant employees and firemen had died due to exposure to radiation. During 1986, about 220 000 people were evacuated from areas surrounding the reactor, and about 250 000 people were relocated subsequently. About 600 000 persons worked, and some still do, in cleaning-up the accident; they are known as 'recovery operation workers' or 'liquidators' (UNSCEAR, 2000).

The radionuclides were released mainly over a period of 10 days after the accident, contaminating vast areas of the Ukraine, Belarus, and the Russian Federation, and trace deposition of released radionuclides was measurable in all countries of the northern hemisphere (UNSCEAR, 1994, 2000). The contamination beyond the 30-km exclusion zone was determined primarily by wind direction. Globally, ^{131}I and ^{137}Cs are the most important radionuclides to be considered, except in the immediate area where deposition reflected the composition of the fuel, since they were responsible for most of the exposure to radiation of the general population. The total releases of ^{131}I and ^{137}Cs in 1996 are estimated to have been 1760 and 85 PBq (1760 and 85×10^{15} Bq; 50% and 30% of the core inventory), respectively (UNSCEAR, 2000).

Three categories of individuals who are likely to have been exposed after the Chernobyl accident to doses of ionizing radiation that could have a measurable effect were the workers involved in the accident (during the emergency or clean-up phase), the inhabitants of evacuated areas and the inhabitants of contaminated areas who were not evacuated. Additionally, individuals residing in the former USSR beyond the heavily contaminated areas and those living in the rest of Europe may have been exposed as a consequence of the accident; however, the dosimetry in these areas is even more complex and unreliable than in those close to Chernobyl.

(i) *Emergency workers*

The emergency workers are those who dealt with the consequences of the accident during the first few days, including staff of the plant, firemen, medical staff and guards. The main exposure was due to relatively uniform external whole-body γ-irradiation and β-irradiation of extensive body surfaces. Acute radiation syndrome was diagnosed in 134 workers (UNSCEAR, 2000), of whom 33 were selected for bone-marrow transplantation. Of these, 13 who were estimated to have received whole-body doses of 5.6–13.4 Gy received bone-marrow transplants (Baranov *et al.*, 1989).

(ii) *Recovery operation workers*

Of particular interest are the 226 000 recovery operation workers who were employed in the 30-km exclusion zone in 1986–87, as it is during this period that the

highest doses were received (UNSCEAR, 2000). The assessments were based largely on group dosimetry or time–activity diaries. The average recorded external doses decreased over time, from about 170 mSv in 1986 to 130 mSv in 1987, 30 mSv in 1988 and 15 mSv in 1989 (Tsyb *et al.*, 1992; Sevan'kaev *et al.*, 1995).

The recovery operation workers received doses not only from external γ- and β-irradiation but also from internal irradiation. Between 30 April and 7 May 1986, direct measurements were made on the thyroids of more than 600 recovery operation workers. The preliminary estimates showed an average dose of 210 mGy. The average effective dose received between June and September 1986 by about 300 recovery operation workers was estimated to have been about 30 mSv. The internal doses from intakes in later years are expected to be much lower: routine monitoring of the ^{137}Cs body burdens indicated average annual doses from ^{137}Cs of 0.1–0.2 mSv in 1987 and 1988 (UNSCEAR, 2000).

(iii) *Evacuees*

Approximately 116 000 individuals were evacuated in 1986 from contaminated areas around the Chernobyl reactor. The effective doses from external exposure for the persons evacuated from the Ukrainian part of the 30-km zone were estimated from measurements performed in this zone and responses to questionnaires from about 35 000 evacuees. The average effective dose from external irradiation for this cohort was estimated to be 17 mSv, with individual values varying from 0.1 to 380 mSv (UNSCEAR, 2000).

In calculations of the effective doses from external irradiation of evacuees from Belarus, it was assumed that 60–80% of the effective dose was contributed by the short-lived radionuclides ^{131}I, ^{132}Te plus ^{132}I, and ^{140}Ba plus ^{140}La, and the contribution from the long-lived radionuclide ^{137}Cs was estimated to be only 3–5%. Overall, it is estimated that about 30% of the people were exposed to effective doses < 10 mSv, about 86% were exposed to doses < 50 mSv, and only about 4% were exposed to doses > 100 mSv, with the average dose estimated to be 31 mSv. The highest average effective dose, about 300 mSv, was estimated to have been received by the populations of two villages located in the 30-km zone (UNSCEAR, 2000).

The average individual and collective doses to the thyroid of evacuees from villages and from the towns of Pripyat and Chernobyl, located in the 30-km zone, are shown in Table 5. The doses from ^{131}I in Pripyat, which were for the most part due to inhalation, were highest for children aged < 1–3 years (mean, about 1.4 Gy), and the average dose for the whole population of this town was 0.2 Gy. The main determinant of the individual dose was found to be the distance of the residence from the reactor (UNSCEAR, 2000).

(iv) *Unevacuated inhabitants of the former USSR*

In the European part of the former USSR, 3% of the land was contaminated after the Chernobyl accident, with ^{137}Cs deposition densities > 37 kBq/m^2. Many people

Table 5. Estimates of doses to the thyroid from intake of ^{131}I by Ukrainian evacuees of towns and villages within a 30-km zone of the Chernobyl reactor

Age at time of accident (years)	Pripyat town			Chernobyl town[a]			Evacuated villages[a]			Total collective dose (person–Gy)
	No. of persons	Arithmetic mean dose (Gy)	Collective dose (person–Gy)	No. of persons	Arithmetic mean dose (Gy)	Collective dose (person–Gy)	No. of persons	Arithmetic mean dose (Gy)	Collective dose (person–Gy)	
< 1	340	2.18	741	219	1.5	329	369	3.9	1 439	2 509
1–3	2 030	1.28	2 698	653	1	653	1 115	3.6	4 014	7 265
4–7	2 710	0.54	1 463	894	0.48	429	1 428	1.7	2 428	4 320
8–11	2 710	0.23	623	841	0.15	126	1 360	0.62	843	1 592
12–15	2 710	0.12	325	846	0.11	93	1 448	0.46	666	1 084
16–18	2 120	0.066	140	650	0.09	59	941	0.39	367	566
> 18	36 740	0.066	2 425	9 488	0.16	1 518	21 794	0.40	8 718	12 661
Total	49 360		8 315	13 591		3 206	28 455		18 475	29 996

From UNSCEAR (2000)

[a] Age distribution of population assumed to be the same as in Pripyat

continued to live in the contaminated territories surrounding the Chernobyl reactor; areas in which the ^{137}Cs deposition density was > 555 kBq/m^2 were considered to be areas of strict control. Initially, 786 settlements inhabited by 273 000 people were considered to be strict control zones. Within these areas, radiation monitoring and preventive measures were taken with the aim of maintaining the annual effective dose within 5 mSv; in 1995, about 150 000 people were living in the areas of strict control (UNSCEAR, 2000). The average effective individual dose received by the inhabitants of these zones was 37 mSv during the first year after the accident (UNSCEAR, 1993). The distribution of the population (a little more than 5 million people) residing in contaminated areas in 1995 according to ^{137}Cs deposition density interval (≥ 37 kBq/m^2) is shown in Table 6. The percentage of the population living in areas with the highest contamination was about 5% in Belarus and the Russian Federation and < 1% in the Ukraine (UNSCEAR, 2000).

Table 6. Distribution in 1995 of inhabitants of areas contaminated by the Chernobyl accident

^{137}Cs deposition density (kBq/m^2)	Population[a]			
	Belarus	Russian Federation	Ukraine	Total
37–185	1 543 514	1 634 175	1 188 600	4 366 289
185–555	239 505	233 626	106 700	579 831
555–1480	97 595	95 474	300	193 369
Total	1 880 614	1 963 275	1 295 600	5 139 489

From UNSCEAR (2000)

[a] For social and economic reasons, some of the populations living in areas contaminated with < 37 kBq/m^2 are also included.

The doses due to internal exposure came essentially from the intake of ^{131}I and other short-lived radioiodines during the first days or weeks after the accident and, subsequently, from intake of ^{134}Cs and ^{137}Cs. Other long-lived radionuclides, notably ^{90}Sr, ^{239}Pu and ^{240}Pu, have so far contributed relatively little to the internal doses, but they will play a more important role in the future. After the Chernobyl accident, about 350 000 measurements of ^{131}I in the thyroid and about 1 million measurements of ^{134}Cs and ^{137}Cs in the whole body were conducted in the three republics by means of γ-radiation detectors placed outside the body (UNSCEAR, 2000).

The main contaminated areas in the Russian Federation are located 150–250 km to the northeast of Chernobyl, and most of the short-lived iodine isotopes had already decayed when the radioactive plume reached this area. Rainfall in the area decreased the concentrations in air and reduced the intake by inhalation. Therefore, the dose to

the thyroid was due primarily to ^{131}I intake with milk and leafy vegetables, and the pattern of doses was similar throughout the region. About 45 000 direct measurements of radioactivity in the thyroid made in May–July 1986 showed a maximum on 16 and 17 May of up to 300 kBq. The doses to the thyroid were estimated in this manner for people in six age groups: < 1, 1–2, 3–5, 7–11, 12–17 and > 18 years. The variations between age groups differed for towns and villages, reflecting not only the age-related metabolism of iodine but also differences in social and nutritional habits. Estimates of doses to the thyroid in contaminated areas of the Russian Federation are presented in Table 7 (UNSCEAR, 2000).

Table 7. Estimated doses to the thyroid of inhabitants of districts of the Russian Federation contaminated by the Chernobyl accident

District/region	Population	Mean thyroid dose (Gy)	Collective thyroid dose (person–Gy)
Bryansk			
Controlled areas[a]	112 000	0.20	22 000
8 contaminated districts	360 000	0.092	33 000
Whole region	1 500 000	0.037	55 000
Tula			
12 contaminated districts	770 000	0.035	27 000
Whole region	1 900 000	0.026	50 000
Orel	870 000	0.017	15 000
Kaluga	1 000 000	0.015	15 000
Total	5 270 000	0.026	135 000

From UNSCEAR (2000)
[a] ^{137}Cs deposition density, > 555 kBq/m^2

(v) *Inhabitants of countries outside the former USSR*

Information on doses received by populations other than those of Belarus, the Russian Federation and the Ukraine is incomplete. The populations of Croatia, Greece, Hungary, Poland and Turkey have been considered in epidemiological studies of thyroid cancer. The average thyroid doses received by these populations were estimated to range from 1.5 to 15 mGy (UNSCEAR, 2000). The collective effective dose from the Chernobyl accident was 600 000 person–Sv (UNSCEAR, 1988), of which 40% is expected to have been received in the former USSR, 57% in the rest of Europe and 3% in other countries of the northern hemisphere (UNSCEAR, 1993).

(*b*) *Southern Ural Mountains*

(i) *History*

The Chelyabinsk region of the southern Ural Mountains was one of the main military production centres of the former USSR and included the Mayak nuclear materials production complex in the closed city of Ozersk. Accidents, nuclear waste disposal and day-to-day operation of the Mayak reactor and radiochemical plant contaminated the nearby Techa River. The period of most releases of radioactive material was 1949–56, with a peak in 1950–51 (UNSCEAR, 2000).

During the first years of the releases, 39 settlements were located along the banks of the Techa River, and the total population was about 28 000. Technical flaws and lack of expertise in radioactive waste management led to contamination of vast areas, and the population was not informed about the releases. The protective measures that were implemented (evacuations, restrictions on the use of flood lands and river water in agricultural production and for domestic purposes) proved to be ineffective, since they were implemented too late. Approximately 7500 people were evacuated from villages near the River between 1953 and 1960 (Akleyev & Lyubchansky, 1994; Degteva *et al.*, 1994).

In 1957, a nuclear waste storage facility in the Chelyabinsk region, near the town of Kyshtym, exploded (the Kyshtym accident) due to a chemical reaction, producing contamination referred to as the East Urals Radiation Trace (EURT). About 273 000 people lived in the contaminated area (Akleyev & Lyubchansky, 1994; UNSCEAR, 2000).

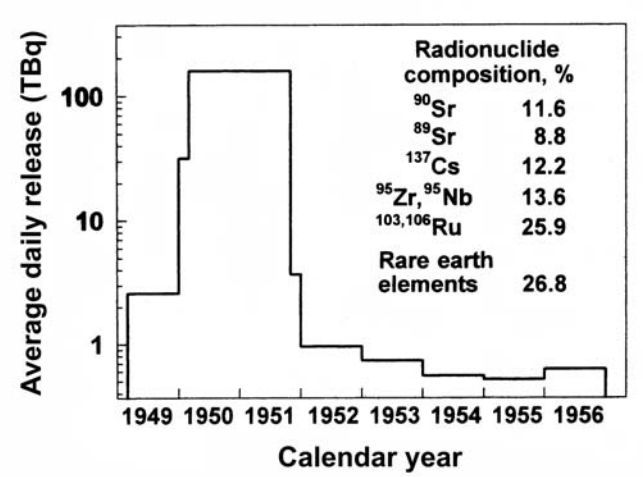
Ten years later, in 1967, after an exceptionally dry summer, the water of the Karachay Lake, an open depot of liquid radioactive waste, evaporated, and a storm transported radionuclides from the dry shores. Eleven thousand individuals were resettled as a result of the Kyshtym accident, of whom 1500 had previously been resettled from the Techa River (Akleyev & Lyubchansky, 1994; UNSCEAR, 2000).

(ii) *Dosimetry*

During 1949–56, 7.6×10^7 m^3 of liquid wastes with a total radioactivity of 100 PBq were released into the Techa–Isset–Tobol river system. The composition of the releases in relation to year of discharge is shown in Figure 1. Large populations were exposed over long periods to external γ-radiation, due largely to ^{137}Cs but also to other γ-emitting radionuclides such as ^{95}Zr, ^{95}Nb and ^{106}Ru present in the water and on the banks of the Techa River. The internal radiation dose was from ingestion of ^{90}Sr and ^{137}Cs over long periods. The doses were estimated as averages for different age groups, assuming that all persons of a specific age living in a specific village in a specific year had accumulated a similar dose (Akleyev & Lyubchansky, 1994; Degteva *et al.*, 1994; UNSCEAR, 2000).

The Techa River dosimetry system was developed in order to estimate the doses received by the persons in the extended Techa River cohort, which consists of approximately 30 000 people, including 5000 who migrated to the area after the period of highest releases. The Techa River offspring cohort consists of 14 000 persons born

Figure 1. Average amount of radioactivity released per day into the Techa River between 1949 and 1956, in relation to year of discharge and radionuclide composition



From Vorobiova *et al.* (1999)
1 TBq = 10^{12} Bq

after 1949 to at least one parent in the extended cohort (Degteva *et al.*, 1996; Kossenko *et al.*, 1997; Degteva *et al.*, 2000a).

The doses from external exposure have been calculated from measurements of external γ-radiation from the River. The internal doses, mainly from ^{90}Sr, have been calculated from the approximately 14 000 whole-body measurements performed since 1970 (Kozheurov & Degteva, 1994; Kossenko *et al.*, 1997; Vorobiova *et al.*, 1999; Degteva *et al.*, 2000a,b).

Recent analyses of the available dosimetry resulted in recalculations of the doses. The major change is increased gastrointestinal doses, due to a larger fraction of short-lived radionuclides and a somewhat smaller contribution from external exposure after revision of data on life-style factors. An important aspect has been reconstruction of what was actually discharged from the Mayak facility. The internal doses are now calculated on the basis of age- and location-specific mean annual intakes of all released radionuclides and individual residence histories. The median dose to the red bone marrow of persons in the extended cohort was 0.21 Gy, and about 50% of the subjects received doses of 0.10–0.50 Gy. The corresponding figures for the distal part of the colon were 0.10 Gy and 0.03–0.20 Gy, respectively. No other tissue except the upper gastrointestinal tract received doses > 0.05 Gy (Degteva *et al.*, 2000a,b).

The external exposure has been found to have been substantially lower because previous calculations were based on the assumption that all residents in a village received the same dose as those living closest to the riverbank (Degteva *et al.*, 1994).

Thus, distance from the River was not taken into consideration. Furthermore, the older calculations overestimated the time spent on the riverbank. External doses are not given in the most recent publication (Degteva *et al.*, 2000a).

(*c*) *Other accidents*

(i) *Windscale*

In October 1957 in Windscale, England, the fuel elements in a graphite-moderated nuclear reactor used to produce plutonium for military purposes caught fire. The fire was detected three days later and, when efforts to extinguish it with carbon dioxide failed, the core was flooded with water. A total of 1.5×10^{15} Bq of radioactive material were released into the environment (Stewart & Crooks, 1958; UNSCEAR, 1993; IARC, 2000), including the radionuclides ^{133}Xe (14×10^{15} Bq), ^{131}I (1.4×10^{15} Bq), ^{137}Cs (0.04×10^{15} Bq) and ^{210}Po (0.009×10^{15} Bq). The total collective effective dose was 2000 person–Sv, including 900 person-Sv from inhalation and 800 person-Sv from ingestion of milk and other foods. Children in the vicinity of the nuclear plant received doses to the thyroid of up to 100 mGy (Burch, 1959; UNSCEAR, 1993; IARC, 2000).

(ii) *Three Mile Island*

The releases of radiation from the accident at the Three Mile Island reactor in Pennsylvania, USA, in March 1979 were caused by failure to close a pressure relief valve, which led to melting of the uncooled fuel. The large release of radioactive material was dispersed to only a minor extent outside the containment building; however, ^{133}Xe (370×10^{15} Bq) and ^{131}I (550×10^{9} Bq) were released into the environment, leading to a total collective dose of 40 person–Sv and an average individual dose from external γ-radiation of 15 μSv. No individual was considered to have received doses to the thyroid of > 850 μSv (UNSCEAR, 1993; IARC, 2000).

The episodes of accidental or non-routine release of radionuclides are summarized in Table 8.

1.1.3 *Routine releases from nuclear installations*

(*a*) *Environmental exposure*

The fraction of electric energy generated by nuclear reactors has grown steadily since their introduction in 1956. By the end of 1997, 437 operating nuclear power reactors and 283 research reactors were in use in 31 countries throughout the world. A total of 285 reactors have now been closed down, 12 are currently under construction, and seven have been planned. The electricity produced by nuclear energy comprised 17% of the electricity generated in the world in 1997 (IAEA, 1997; UNSCEAR, 2000).

The nuclear fuel cycle includes mining and milling of uranium ore, conversion into fuel material, production of fuel elements, energy production and storage and reprocessing. The doses to individuals from routine releases from nuclear installations

Table 8. Characteristics of accidental or non-routine releases of radionuclides

Site	Approximate quantity released	Date	Collective effective dose	Individual effective dose
Chernobyl, Russian Federation	^{131}I, 1760 PBq; ^{137}Cs, 85 PBq	1986	600 000 person–Sv	31 mSv for evacuees
Techa River, southern Urals, Russian Federation	~ 100 PBq	1949–56	Not estimated	Average bone-marrow dose for those living along the River, 21 mSv
Kyshtym, southern Urals, Russian Federation	74 PBq	1957	Not estimated	Not estimated
Karachay Lake, southern Urals, Russian Federation	22 TBq	1967	Not estimated	Not estimated
Windscale, United Kingdom	^{131}I, 1.4 PBq	1957	2000 person–Sv	Dose to the thyroid: average, 5–125 μSv; maximum, 5 mSv
Three Mile Island, USA	^{131}I, 550 GBq; ^{133}Xe, 370 PBq	1979	40 person–Sv	External γ: average, 15 μSv; maximum 850 μSv

From UNSCEAR (1993); Akleyev & Lyubchansky (1994); Burkart (1996); Degteva *et al.* (2000a); UNSCEAR (2000)
P, peta = 10^{15}; T, tera = 10^{12}; G, giga = 10^{9}

vary considerably according to the installation and with time, but the most important determinant of dose is the distance from the release (UNSCEAR, 2000).

Airborne and liquid radioactive releases from nuclear reactors during routine operations have been reported with substantial completeness. Models for calculating the actual doses from the releases for each radionuclide and combination of radionuclides are complex. Factors to be taken into consideration include the geographical location of the reactor, population density and distribution, food production, consumption habits, environmental pathways, dilution of radionuclides, composition of radionuclides and type of reactor. The concentrations of radionuclides are generally not measurable, except close to the nuclear plant, and then only for a limited number of radionuclides. The calculation of individual and collective doses is therefore dependent on modelling of atmospheric and aquatic transport and environmental transfer before application of dosimetric models (UNSCEAR, 2000).

In a recent report (UNSCEAR, 2000), the total collective dose for the period 1990–94 was estimated to be 490 person–Sv, which represents a 25% increase over the preceding five-year period and corresponds to the approximate 25% increase in energy production during that time. The collective effective dose for a given reactor is described in person–sieverts per unit of electric energy generated. For the period 1990–94, that value was calculated to be 0.43 person–Sv/GW–year; noble gases and tritium (^{3}H) (airborne and liquid) contributed approximately 25% and 19%, respectively, while ^{131}I contributed 0.0002 person–Sv/GW–year (Table 9). The calculations were made under the assumption that the population density was 20/km^2 within 2000 km and 400/km^2 within 50 km of the release. The annual effective doses of most individuals throughout the world were calculated on the basis of the data above and, depending on the type of reactor, resulted in estimates of 0.4–10 μSv.

The change in releases over time and the collective effective doses of different nuclides are shown in Figure 2. The total collective dose has been stable since 1970–74, whereas the quantity of electrical energy generated increased between 1970 and 1994. The collective effective dose from ^{131}I has diminished dramatically after a peak in 1975–79 (UNSCEAR, 2000).

(*b*) *Occupational exposure* (see also section 1.2)

Occupational exposure in nuclear installations varies significantly, depending on the type, size and age of the reactor. Other factors that affect the radiation dose are changes in operating routines, piping, shielding and reactor water chemistry. In contrast to environmental exposure, in which internal exposure is the main component of dose, occupational exposure consists almost entirely of external exposure to γ-radiation and occurs mostly during scheduled maintenance and/or refuelling.

Occupational exposure at reactor sites, regardless of the type, has decreased steadily in the past few years throughout the world. Difficulties in comparing doses between countries arise due to differences in reporting, e.g. in some cases, only workers

Table 9. Normalized collective effective doses from radionuclides released from nuclear reactors, 1990–94

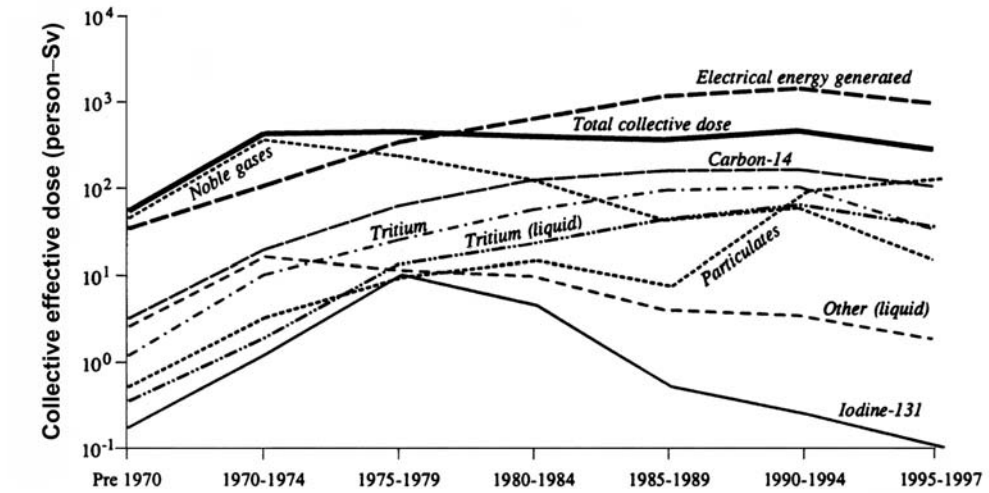
Reactor type	Electrical energy generated (%)	Collective effective dose per unit electrical energy generated (person–Sv/GW–year)						
		Airborne effluents					Liquid effluents	
		Noble gases	^{3}H	^{14}C[a]	^{131}I	Particulates	^{3}H	Other
PWR	65.04	0.003	0.005	0.059	0.0001	0.0004	0.014	0.006
BWR	21.95	0.15	0.002	0.14	0.0002	0.36	0.0006	0.014
GCR	3.65	1.44	0.010	0.38	0.0004	0.0006	0.14	0.17
HWR	5.04	0.23	1.4	0.43	0.0001	0.0001	0.32	0.043
LWGR	4.09	0.19	0.05	0.35	0.002	0.028	0.007	0.002
FBR	0.24	0.042	0.10	0.032	0.00009	0.024	0.0012	0.016
Weighted average		0.11	0.075	0.12	0.0002	0.080	0.031	0.016
Total					0.43			

From UNSCEAR (2000)

[a] Local and regional components only

BWR, boiling water reactor; FBR, fast breeder reactor; GCR, gas-cooled and graphite-moderated reactor; HWR, heavy water-cooled and -moderated reactor; LWGR, light water-cooled and graphite-moderated reactor; PWR, light water-moderated and -cooled pressurized reactor

Figure 2. Local and regional collective effective doses from average annual releases of radionuclides from reactors



From UNSCEAR (2000)

The increasing trend in electrical energy generated is indicated on the scale on the left, in units of GW-year.

with measurable doses are included, which affects the average dose of the workforce. The annual average collective effective dose for 1990–94 was calculated to be 900 person–Sv, which was lower than the dose of 1100 person–Sv calculated for the preceding period. In 1990–94, the annual effective dose among monitored workers, averaged over all reactors, was 1.4 mSv, and that for measurably exposed workers was 2.7 mSv (UNSCEAR, 2000).

1.1.4 *Dosimetry of radon-222*

Studies of underground miners indicate that the rate of mortality from lung cancer is related directly to the concentration of radon and its decay products in the air of uranium and other hard-rock mines. Exposure was reduced in many mines after recognition that breathing radon gas and its decay products is an occupational risk. Other important sources of exposure to radon have since been recognized; in particular, it has been shown that radon can accumulate in appreciable concentrations in homes, offices and other buildings where air exchange with outdoor air is restricted. Exposures from residential or indoor radon have been studied extensively around the world (Committee on the Biological Effects of Ionizing Radiations (BEIR IV), 1988; IARC, 1988; ICRP, 1993a; Committee on Health Risks of Exposure to Radon (BEIR VI), 1999; Lubin *et al.*, 1995a, 1997; UNSCEAR, 2000).

The dosimetry of radon, i.e. the dose to the bronchial epithelium, is complex and depends on factors such as the concentrations of radon and radon decay products in inspired air, aerosol factors such as the size distribution of inhaled particles and the attachment of radioactive particles to the aerosol and the distribution of radionuclides. The physiological factors include lung morphology, depth of target cells, amount of air moving through the lung per time unit, particle deposition fraction, mucus thickness and transport rate through mucus. These considerations are dealt with comprehensively in several publications (Committee on the Biological Effects of Ionizing Radiations (BEIR IV), 1988; Committee on Health Risks of Exposure to Radon (BEIR VI), 1999), and only a brief overview is given here.

The risk for lung cancer of miners exposed to radon is expressed in units of working-level months (WLM), whereas lung cancer risks in homes are evaluated in terms of Bq/m^3 or time-weighted average Bq/m^3. Living in an atmosphere of 1 pCi/L (37 Bq/m^3) for one year is comparable to 0.14 WLM, and 1 WLM results in a dose to the lung epithelium of about 6 mGy. Thus, a typical indoor concentration of radon of about 40 Bq/m^3 would result in a yearly dose to the lung of about 0.84 mGy (Committee on the Biological Effects of Ionizing Radiations (BEIR IV), 1988; UNSCEAR, 2000). Assuming a radiation weighting factor of 20 for α-particles would result in an equivalent dose to the lung of about 17 mSv/year. As described below, the dosimetry and conversions are complex and not entirely straightforward, and there remains some debate about the best dosimetry models and assumptions to be made.

Nevertheless, it is clear that the presence of radon at sufficiently high levels in residential areas poses an important risk for lung cancer.

(*a*) *Decay of radon and its progeny*

^{226}Ra is the immediate parent of ^{222}Rn, and radium is the fifth progeny of ^{238}U, which has a half-life of 4.5 thousand million years, which is similar to the age of the earth. A typical concentration of uranium in ordinary soil is 20 Bq/kg. ^{226}Ra has a half-life of 1600 years. The decay scheme of ^{222}Rn starts with ^{226}Ra and ends with stable ^{206}Pb. The first four progenies of radon (^{222}Rn), ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po, have half-lives that are shorter (all < 30 min) than the 22-year half-life of the fifth progeny, ^{210}Pb. Nearly 90% of the decay energy of the short-lived progenies occurs by α-particle emission, even though two of these decay by β-particle emission with associated γ-rays. Thus, under most circumstances, only the short-lived α-particle-emitting progenies are of consequence in the respiratory dosimetry of the radon chain. It should be mentioned that radon is an inert gas and ^{222}Rn has a half-life of 3.8 days, which is much longer than the time of ventilation (a few seconds). Because inhaled radon is largely exhaled again, it contributes negligibly to the dose to the lung, and it is only the short-lived progeny that are of concern. A typical concentration of radon in air over average soil is 4 Bq/m^3 (Committee on the Biological Effects of Ionizing Radiations (BEIR IV), 1988).

(*b*) *Quantities and units*

Holaday *et al*. (1957) introduced the working level (WL) as a convenient measure of the concentration of radon progeny in the air of uranium mines, which can be used as a measure of exposure. WL was defined as the combination of the short-lived progeny of radon (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) in 1 L of air, under ambient temperature and pressure, that results in the ultimate emission of 1.3×10^5 MeV α-particle energy. This is approximately the total amount of energy released over a long period by the short-lived decay products in equilibrium with 100 pCi (3.7 Bq) of ^{222}Rn.

Only the short-lived progeny of radon are included in the definition of WL because they contribute most of the dose to the lung. The dose from β-particles is small, and the α-particles from radon itself are unlikely to be emitted within the body because almost all inhaled radon is exhaled. Most of the ^{210}Pb (22-year half-life) and subsequent progeny are probably eliminated from the body before they decay, although low levels of ^{210}Pb can be measured in heavily exposed miners many years later (Committee on the Biological Effects of Ionizing Radiations (BEIR IV), 1988; Committee on Health Risks of Exposure to Radon (BEIR VI), 1999).

The SI unit for the potential α-energy concentration of radon decay products in air is J/m^3, where 1 J/m^3 = 6.24×10^{15} eV/L of air. 1 WL = 1.3×10^5 MeV/L = 2.08×10^{-5} J/m^3. The WLM was developed to account for both the duration and the level of exposure. It is defined as the product of the WL times the duration of exposure, i.e. during one month of 170 working hours. The unit WLM is equal to 170 WLh, i.e. exposure of 1 WL for 170 h (Committee on the Biological Effects of Ionizing Radiations

(BEIR IV), 1988). Activity is defined as the number of radioactive transformations of a radionuclide over unit time and is expressed in becquerels.

The concentration of radon decay products in indoor air is often expressed in terms of the 'equilibrium equivalent concentration' of radon. It corresponds to the activity concentration of radon for which the short-lived decay products in equilibrium with the parent have the same potential α-particle energy as radon itself. In practical terms, ventilation and deposition of radon decay products on surfaces are such that radioactive equilibrium is rarely reached. In order to account for this, an equilibrium factor (F) is used. The equilibrium factor is defined as the ratio of the potential α-particle energy concentration of the decay products to the corresponding concentration if they were in radioactive equilibrium with radon.

The equilibrium equivalent concentration of radon decay products in indoor air is expressed in units of Bq/m^3 as $F \times$ the activity concentration of radon (IARC, 1988).

(*c*) *Dose*

The relationship between exposure to radon progeny, whether measured as WLM or estimated as Bq/m^3, and the dose of α-energy delivered to cells in the respiratory tract, considered as targets for carcinogenesis, is extremely complex and depends on both biological and non-biological factors. Since the dose of α-energy delivered to the target cells in the lungs cannot be measured directly, models are used to simulate the sequence of events, from inhalation of radon progeny to cellular injury. These complex models generally include biological factors, such as airway geometry, mucociliary clearance, particle deposition, ventilation pattern and location of the target cells in the lung. Physical factors of importance are the amount of air inhaled, the aerosol size distribution and the proportion of progeny not attached to particles. Factors for converting exposure to an absorbed radiation dose can be calculated by using dosimetric models of the respiratory tract, but the range of published conversion factors is wide (Committee on Health Risks of Exposure to Radon (BEIR VI), 1999).

The ratio of the dose of α-energy per unit exposure for a particular population group (men, women, children, infants), as given by the radon concentration, to the dose per unit radon concentration for miners is given by the K factor, which is defined as:

K = [dose (home)/exposure (home)]/[dose (mine)/exposure (mine)].

The K factor includes diverse environmental and physiological factors, and use of the double ratio simplifies the risk assessment for indoor radon. The dose-rate per unit radon concentration for miners is 7.0 nGy/h per Bq/m^3 . The National Research Council (1991), in its analysis of lung dosimetry, found a K factor of 0.75 for males and females and somewhat higher values for children and infants. The Committee on Health Effects of Exposure to Radon (BEIR VI) (1999) of the National Research Council determined that the median K factor in new computations was closer to 1, implying that the correction factor necessary for extrapolating data for mines to data for homes was not large (Committee on Health Risks of Exposure to Radon (BEIR VI), 1999).

The latest report of UNSCEAR (2000) on natural activity summarized much of what is now accepted about exposure and dose to the lung for miners and from indoor radon concentrations: the studies of mines and homes give the same dose factor.

1.2 Occupational exposure

1.2.1 *Monitoring*

Differences in the results of monitoring of exposure of workers to radiation reflect changes over time in monitoring practices and techniques and simultaneous use of different methods to monitor the same exposure (UNSCEAR, 1993). Environmental or area monitoring, with air sampling or radiation monitoring devices at the entrance and exit of work areas, may provide information about the presence of radiation sources in the environment (UNSCEAR, 2000). Work surfaces may be routinely swabbed in order to obtain information about environmental contamination. When such contamination is detected, workers may be evaluated for internal exposure with mouth or nose swipes, estimates of contamination of clothing and/or skin and quantitative exposure assessments. When an incident has led to internal deposition of radioactive material, e.g. from a puncture wound while handling radioisotopes or accidental inhalation of volatile radioactive material, the exposure will usually be assessed by repeated individual monitoring

It is now quite common for workers with potential internal exposure to radiation to undergo routine (e.g. annual) monitoring, in order to ensure that the working conditions meet regulatory requirements (ICRP, 1991, 1994a; UNSCEAR, 2000). Task-related monitoring is sometimes undertaken to support decisions and actions to improve working conditions and to optimize protection. In general, individual monitoring involves more workers than are strictly necessary to meet regulatory requirements, and only a fraction of those monitored receive measurable doses (UNSCEAR, 2000).

(*a*) *Internal exposure*

Three main methods are used to assess individual internal exposure to radionuclides: personal air sampling, i.e. in the breathing zone of individual workers; in-vitro monitoring by analysis of biological materials; and in-vivo measurement of internal contamination by γ-spectroscopy. Personal air sampling may provide a measure of exposure in terms of the time-integrated air concentration of radionuclides in the breathing zone of individual workers. This is the only available method for routine monitoring of radon (UNSCEAR, 2000).

In-vitro monitoring involves measurement of radioactivity in concentrated or desiccated body fluids such as blood, urine or faeces. This type of monitoring has been used for many decades in the nuclear industry. For α- and β-particle emitters such as readily measurable, highly soluble radionuclides like ^{3}H, in-vitro monitoring is often the most sensitive method.

In-vivo monitoring of radionuclides that emit highly penetrating X- or γ-radiation, by counting of the whole body, thorax, skeleton and thyroid of a worker with a γ-ray spectrometer in a heavily shielded room, also gives the energy spectrum of the radionuclides. This technique was first used in the early 1960s. The two types of whole-body counters that have been used routinely are liquid scintillation counters, primarily in earlier periods, and solid crystal systems. When an internally deposited radionuclide does not emit X- or γ-radiation during radioactive decay, in-vivo monitoring may rely on detection of γ-rays emitted by associated radionuclides, such as γ-ray-emitting ^{241}Am, which is often deposited with plutonium. In-vivo monitoring of internal deposition of β-particle emitters, such as ^{32}P, may be based on the detection of the *bremsstrahlung* produced by the β-particles (McCunney *et al.*, 1999). In many situations, a combination of methods is used.

(*b*) *Accuracy and reliability of measurements*

The dose from intake of radioactive materials usually cannot be measured with the same degree of accuracy as that from external radiation. In particular, personal air sampling may not be adequate to estimate the annual intakes of individual workers exposed at the levels usually encountered in occupational settings. In a study of long-term low-level exposure of workers in nuclear fuel reprocessing, dose assessments obtained by static air sampling, personal air sampling and analysis of biological samples *in vitro* were compared. Personal air sampling provided a dose estimate that was about an order of magnitude higher that that deduced from static air sampling. For the group as a whole, there was reasonable agreement between the cumulative doses derived from biological sampling (23 mSv) and personal air sampling (30 mSv) over a seven-year period; however, at the individual level there was no correlation between the two sets of data (Britcher & Strong, 1994).

A number of factors may introduce uncertainty and/or bias into estimates of radiation dose from internally deposited radionuclides. One is uncertainty about the time of intake, which must be known in order to determine the organ burden from an in-vitro or in-vivo monitoring result. The most accurate assessment of organ or body burden can be made when an incident is known to have occurred and measurements were made *in vitro* and *in vivo* over time. In this case, a biological half-time can be obtained from the actual data. However, monitoring is usually conducted routinely (often annually), and organ burdens are difficult to deduce. When the time of intake is unknown, assumptions must be made about the pattern of activity between the time of first employment and the first measurement, about the organ burden between the time of the last measurement and the termination of employment and about the organ burden between measurements. If there is continuous intake of a radionuclide, it is reasonable to assume an average organ burden during the period between measurements, but this would not be appropriate if acute intake had occurred, and the method of integration must account for such intakes. In addition, accumulated activity and corresponding dose must be apportioned by year.

Internal dose can be calculated after γ-ray spectrometry *in vivo* from measurements of radioactive decompositions. When dose estimates derived from whole-body counting methods are used, the estimates of internal radiation dose may be erroneous, because of inhomogeneous distribution of radionuclides in the body and inappropriate assumptions about the ratios of radioisotopes ingested (e.g. the ratio of americium to plutonium). In addition, the detection limits of whole-body counters have changed over time and vary by radioisotope. In order to estimate internal organ doses from analyses of urine or any other biological sample, the amount of an isotope in a reference volume of the sample must be correlated with the amount in the body or in a specific organ. To achieve this, the activity of a nuclide eliminated per unit time must be determined, the activity in organs of interest must be inferred from the eliminated activity per unit time and the organ activity must be integrated over time to determine the cumulated organ burden and the total dose. Uncertainty in these estimates results from differences in excretion rates between individuals and within an individual over time and difficulties in characterizing the biological distribution of radionuclides in the body. Methods for calculating internal doses from in-vitro monitoring data follow from recommendations issued by the ICRP (1979, 1980, 1981, 1986). Estimates of internal exposure derived from air monitoring data are based on the integration over time of measured air concentrations of radioactive materials. Models are then used to estimate internal deposition from inhalation or ingestion of the radionuclide. While each of these internal dosimetry methods provides useful information for evaluating, and controlling, internal exposure to radiation, uncertainties in the estimates of the ensuing effective doses may be substantial and may be an important limitation when these data are to be used in epidemiological analyses. The accuracy of occupational exposure estimates has been assessed in several comparisons of estimates of body burden from analysis of urine samples and tissue taken at autopsy. Discrepancies were found: for example, urine-based assessments overestimated the plutonium body burden by approximately fivefold (Lagerquist *et al*., 1969; Heid, 1983).

1.2.2 *Exposed populations*

Workers employed in a number of industries have potential internal exposure to natural or man-made sources of ionizing radiation. Significant exposure can occur in mining of radioactive ores (uranium, mineral sands), and exposure to radon can occur in these and other mines, in caves open to tourists and in some above-ground workplaces. The luminizing industry provided an appalling example of the occupational hazards of radium. At present, however, occupational situations in which there is significant internal exposure to radionuclides are rare, and exposures have generally been decreasing. Activities in which exposure to internal radiation from man-made radionuclides may still be significant are the production of nuclear weapons, some situations in nuclear medicine for both diagnostic and therapeutic purposes and the nuclear fuel cycle.

Many of the data on occupational exposure cited below were taken from UNSCEAR (2000). Such data have been recorded systematically in various countries by national authorities and have been collected through an UNSCEAR questionnaire that is distributed throughout the world. In some cases, the data are supplemented by other — usually published — information; for example, the databank of the Organization for Economic Co-operation and Development (OECD)/Nuclear Energy Agency was used as an additional source of information on the nuclear power industry (OECD, 1996, 1998).

The generation of nuclear energy involves mining and milling of uranium, uranium enrichment, reactor fuel fabrication, reactor operation, nuclear fuel reprocessing, waste handling and disposal and research and development. For each main stage of this fuel cycle, except for treatment and disposal of solid radioactive waste, extensive data on occupational exposure and dose distributions are available. In the future, the decommissioning of nuclear reactors will become an important stage (UNSCEAR, 2000).

(*a*) *Uranium mining and milling*

Uranium mining and milling involve underground or open-pit removal of uranium ore, crushing and grinding of raw ore, followed by chemical leaching, separation of uranium from the leachate and precipitation as ‘yellowcake’. In the period 1990–92, worldwide uranium production by underground mining decreased from 55% to 45%, production by open-pit mining increased from 38% to 44%, while production by *in situ* leaching increased from 6% to 9%. Internal exposure during mining and milling of uranium ores may occur through inhalation of radon gas and radionuclides in ore dust. More internal exposure occurs in underground mines than in open mines, where inhalation of radioactive dust is probably the main source of internal exposure (UNSCEAR, 2000).

Urine was collected between 1950 and 1953 from 249 uranium mill workers in the USA and analysed for the presence of uranium. The concentrations were 0–3.0 μg/L for 80 workers, 3.1–9.0 μg/L for 116 workers, 9.1–30 μg/L for 46 workers and 30–160 μg/L for seven workers (Archer *et al.*, 1973).

The uranium mine at Rössing in Namibia consists of an open-pit area and a uranium milling plant. The average background radiation dose (excluding radon progeny) of all the miners was reported to be 1.8 mSv/year. Fourteen of these miners and six controls were chosen for assessment of uranium exposure by analysis of 24-h urine samples. The urinary concentration of ^{238}U (9.57 ± 7.9 mBq/L) was sixfold greater than the control level (1.5 ± 1.1 mBq/L; $p < 0.001$) (Zaire *et al.*, 1996).

Worldwide exposure during uranium mining and milling is summarized in Tables 10 and 11 (UNSCEAR, 2000).

Table 10. Occupational exposure in uranium mining worldwide

Period[a]	Annual amount of uranium ore extracted (thousand tonnes)	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	52	240	1300	5.5
1980–84	64	310	1600	5.1
1985–89	59	260	1100	4.4
1990–94	39	69	310	4.5

From UNSCEAR (2000)

[a] Data are annual averages over the periods indicated. For 1990–94, the worldwide estimates are extrapolated from the total amount of uranium mined worldwide relative to the sum of the total for which an estimate was made.

Table 11. Occupational exposure in uranium milling worldwide

Period[a]	Annual amount of uranium ore refined (thousand tonnes)	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	53	12	124	10.1
1980–84	64	23	117	5.1
1985–89	58	18	116	6.3
1990–94	39	6	20	3.3

From UNSCEAR (2000)

[a] Data are annual averages over the periods indicated. The worldwide estimate is based on the assumption that the amount of uranium ore refined is equal to the amount mined.

(*b*) *Uranium enrichment and conversion*

During uranium conversion, U_3O_8 from the milling process is reduced to UO_2 by reduction with hydrogen, and the UO_2 is then converted to UF_4 by addition of hydrofluoric acid, and subsequently to UF_6 with fluorine. The UF_6 is then enriched in ^{235}U (to about 3%; natural uranium contains about 0.7% ^{235}U) by gaseous centrifugation. After enrichment, UF_6 is reconverted into UO_2 for fuel fabrication. During these processes, occupational exposure is mainly to external radiation, but workers may be exposed to internal irradiation during maintenance work or in the event of leaks.

The Y12 plant in Oak Ridge, TN (USA), was built in 1943 for enrichment of uranium for nuclear weapons. In 1947, its activities shifted to fabrication of weapons parts and nuclear research. Exposure to airborne uranium dust was the major concern at this plant, in particular during the reduction of UF_4 to metal, casting of the metal

and extraction of the UF_4. Estimates of dose equivalents of uranium delivered to the lungs of 3490 workers were obtained from urine analysis and in-vivo counting of internally deposited uranium. For cumulative doses of internal α-irradiation, values of more than 0.3 Sv were found for about 5% of these workers, 0.1–0.3 Sv for about 25%, 0.01–0.1 Sv for about 50% and < 0.01 Sv for about 20%. These levels are at least an order of magnitude lower than those estimated for uranium miners (Checkoway *et al.*, 1988).

A group of 991 workers employed between 1943 and 1949 at the ceramics plant of Linde Air Products (Buffalo, NY, USA) were monitored for internal exposure to uranium. The plant was mainly involved in uranium processing and conversion to UF_4, and experimental work was done on conversion of UF_6 to UO_3. The potential doses to the lung were estimated from the concentration of uranium in the urine of these workers: 212 workers had an annual lung dose < 10 mSv, 402 workers had doses of 10–100 mSv, and 377 workers had doses > 100 mSv (Dupree *et al.*, 1987).

During four five-year periods between 1975 and 1994, 11 000, 4300, 5000 and 12 600 workers were monitored and found to have total annual collective effective doses of 5.3, 0.78, 0.43 and 1.28 person–Sv, respectively. These doses would correspond to average annual effective doses per worker of 0.46, 0.18, 0.08 and 0.10 mSv in these periods, respectively. It should be noted that uranium is enriched and converted in only seven countries and that the worldwide data should be considered rough estimates, as not all these countries provided data for each of the periods (UNSCEAR, 2000).

(*c*) *Reactor fuel manufacture*

Depending on the reactor type, four types of uranium fuel are used: un-enriched uranium metal fuel (used in gas-cooled Magnox reactors), low-enriched UO_2 fuel (used in advanced gas-cooled, graphite-moderated reactors and in light water-moderated and cooled reactors), un-enriched UO_2 fuel (heavy water-cooled and -moderated reactors) and mixed UO_2–PuO_2 fuel (used in light-water and fast-breeder reactors). The main source of exposure during fuel manufacture is uranium, because most decay products are removed during enrichment and conversion. Exposure is from external γ-irradiation and intake of airborne radionuclides. Exposure during reactor fuel manufacture is summarized in Table 12. Despite an approximately threefold increase in the volume of fuel produced, the collective dose has been relatively constant over time. It follows that the collective dose per amount of fuel or per unit energy produced has decreased considerably over the past 20 years (UNSCEAR, 2000).

(*d*) *Reactor operations*

Nuclear reactors for the generation of electrical energy are characterized by their coolant system and moderator: light water-moderated and -cooled pressurized or boiling-water reactors, heavy water-moderated and -cooled reactors, gas-cooled, graphite-moderated reactors and light water-cooled, graphite-moderated reactors. The

Table 12. Occupational exposure in nuclear fuel manufacture worldwide

Period[a]	Annual production of fuel (thousand tonnes of uranium)	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	3.6	20	36	1.8
1980–84	6.1	21	21	1.0
1985–89	9.7	28	22	0.8
1990–94	11.3	21	22	1.0

From UNSCEAR (2000)

[a] Data are annual averages over the periods indicated.

moderator material is used to slow down the fast neutrons generated during the fission of uranium. At the end of 1997, there were 437 nuclear power stations operating worldwide, with a capacity of about 352 GWe (net gigawatts electric), supplying approximately 17% of the total electric energy generated in the world and accounting for about 6% of the world's energy consumption. Over 300 of the nuclear power stations (three-quarters of the total number) are light-water reactors (UNSCEAR, 2000).

Occupational exposures during normal reactor operation can vary considerably, depending on the size, type and age of the reactor, reactor water chemistry and operating procedures. External exposure to γ-radiation is the most significant component of occupational exposure around nuclear reactors. Workers at heavy-water reactors may have internal exposure to radionuclides, particularly ^{3}H, as deuterated water is used as both the coolant and the moderator. In these reactors, neutron activation of deuterium produces a significant amount of ^{3}H (UNSCEAR, 2000).

Workers at the Dounreay Establishment (Atomic Energy Authority) in the United Kingdom, a prototype fast reactor in operation since 1973, were monitored for internal exposure to radionuclides by a combination of personal and static air sampling, excretion monitoring and in-vivo counting. In addition to electricity production, the reactor provides facilities for testing materials and developing advanced fuels. Plants for fuel reprocessing and manufacture of fuel elements are also located on this site. During 1987, the workforce (n = 1778) received a total dose of 3.93 person–Sv, of which 3.21 person–Sv were from external radiation and 0.72 person–Sv (committed effective dose equivalent) from internal radionuclides. In 1988 (workforce, 1702 persons), these values were 4.11 (total), 3.21 (external) and 0.90 person–Sv (internal exposure). The highest internal doses were received by workers in waste management, engineering support and analytical services. During these two years, the exposure of none of the workers exceeded the self-imposed target of 15 mSv (committed effective dose equivalent) (Smith *et al.*, 1991).

Internal exposure to radionuclides was measured for workers at nuclear power stations in France, as part of a routine medical control programme. Between 1980 and 1985, the number of urine analyses increased from about 2200 to 7300 per year, and < 0.5% of the samples were active. The major isotopes detected were ^{58}Co, ^{60}Co, ^{131}I and ^{137}Cs, with detection limits in routine analyses of 0.6, 0.5, 0.7 and 0.8 Bq, respectively (Kwadow & Chevalier, 1988).

Data on occupational exposures at reactors worldwide are given in Table 13. The annual effective dose of monitored workers, averaged over all reactors, decreased from 4.1 to 1.4 mSv over the periods 1975–79 and 1990–94, while the energy production increased by more than fourfold. Accordingly, the dose per unit of energy produced has decreased considerably over the past two decades (UNSCEAR, 2000).

Table 13. Occupational exposure in normal reactor operations worldwide

Period[a]	Total no. of reactors	Average amount of energy generated (GW–year)	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	190	5	150	600	4.1
1980–84	280	100	290	1000	3.5
1985–89	400	190	430	1100	2.5
1990–94	421	230	530	900	1.4

From UNSCEAR (2000)

[a] Data are annual averages over the periods indicated.

(*e*) *Fuel reprocessing*

Irradiated spent fuel from nuclear power stations on a commercial scale is reprocessed only in France and the United Kingdom, while smaller facilities are in operation in India, Japan and the Netherlands. The Russian Federation has been reprocessing fuel for reactors developed in that country. The process involves dissolution of the spent fuel elements in acid, followed by chemical separation of uranium and plutonium from the fission products and other compounds in the fuel. At the time of reprocessing, the fuel still contains high levels of radioactive materials, and heavy shielding and remote operations are required for adequate protection of workers. Both in France and the United Kingdom, the average annual effective dose of monitored workers decreased steadily over the four five-year periods between 1975 and 1994, from 4.03 to 0.36 mSv and from 8.31 to 2.03 mSv, respectively. The Japanese data showed an increase during the period 1975–89 from 0.44 to 0.98 mSv, and a decrease to an average of 0.32 mSv over the following five years. Data from the Russian Federation over the period 1990–94 showed an average annual effective dose of 2.82 mSv (UNSCEAR, 2000).

(*f*) *Waste management*

Only a very small portion of the radioactive waste from the nuclear fuel cycle has been moved to final repositories, but the doses associated with waste management are of increasing importance. In 1993, the cumulative total amount of spent fuel arising from all types of reactors was estimated to be about 145 000 tonnes of heavy metal, whereas the storage capacity at reactors is about 59 000 tonnes of heavy metal. The exposure associated specifically with waste management is not readily known, and the doses associated with waste management have been subsumed into data for reactor operation, reprocessing and research (UNSCEAR, 2000).

(*g*) *Research in the nuclear fuel cycle*

Worldwide occupational exposure arising in nuclear research is summarized in Table 14. The annual number of workers remained relatively constant, and the annual collective dose decreased from 170 to 90 person–Sv in the two decades between 1975 and 1994. Accordingly, the average annual effective dose per worker decreased from 1.4 to 0.8 mSv. Among the workers monitored, the proportion exposed to > 15 mSv decreased from 4% to 1% during the same period (UNSCEAR, 2000).

Table 14. Occupational exposure in research in the nuclear fuel cycle worldwide

Period[a]	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	120	170	1.4
1980–84	130	150	1.1
1985–89	130	100	0.8
1990–94	120	90	0.8

From UNSCEAR (2000)
[a] Data are annual averages over the periods indicated.

Worldwide annual exposure during the commercial nuclear fuel cycle is summarized in Table 15. The number of workers monitored increased to 880 000 up to 1989 and then decreased to 800 000, largely because of a substantial reduction in the number of workers in the mining sector. This also explains the decrease in collective effective dose during the period 1985–94. At present, about 530 000 workers (65%) are involved in reactor operation. Table 15 also shows the downward trend in the collective effective dose per unit nuclear energy generated, with an approximately twofold reduction over the periods studied. The trends in numbers of monitored workers and doses of workers in the various sectors of the nuclear fuel cycle are illustrated in Figure 3 (UNSCEAR, 2000).

Table 15. Average annual occupational exposure in the commercial fuel cycle worldwide

Period[a]	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Total annual collective effective dose per unit nuclear energy generated (person–Sv per GW–year)	Average annual effective dose (mSv)
1975–79	560	2300	20	4.1
1980–84	800	3000	18	3.7
1985–89	880	2500	12	2.9
1990–94	800	1400	9.8	1.8

From UNSCEAR (2000)
[a] Data are annual averages over the periods indicated.

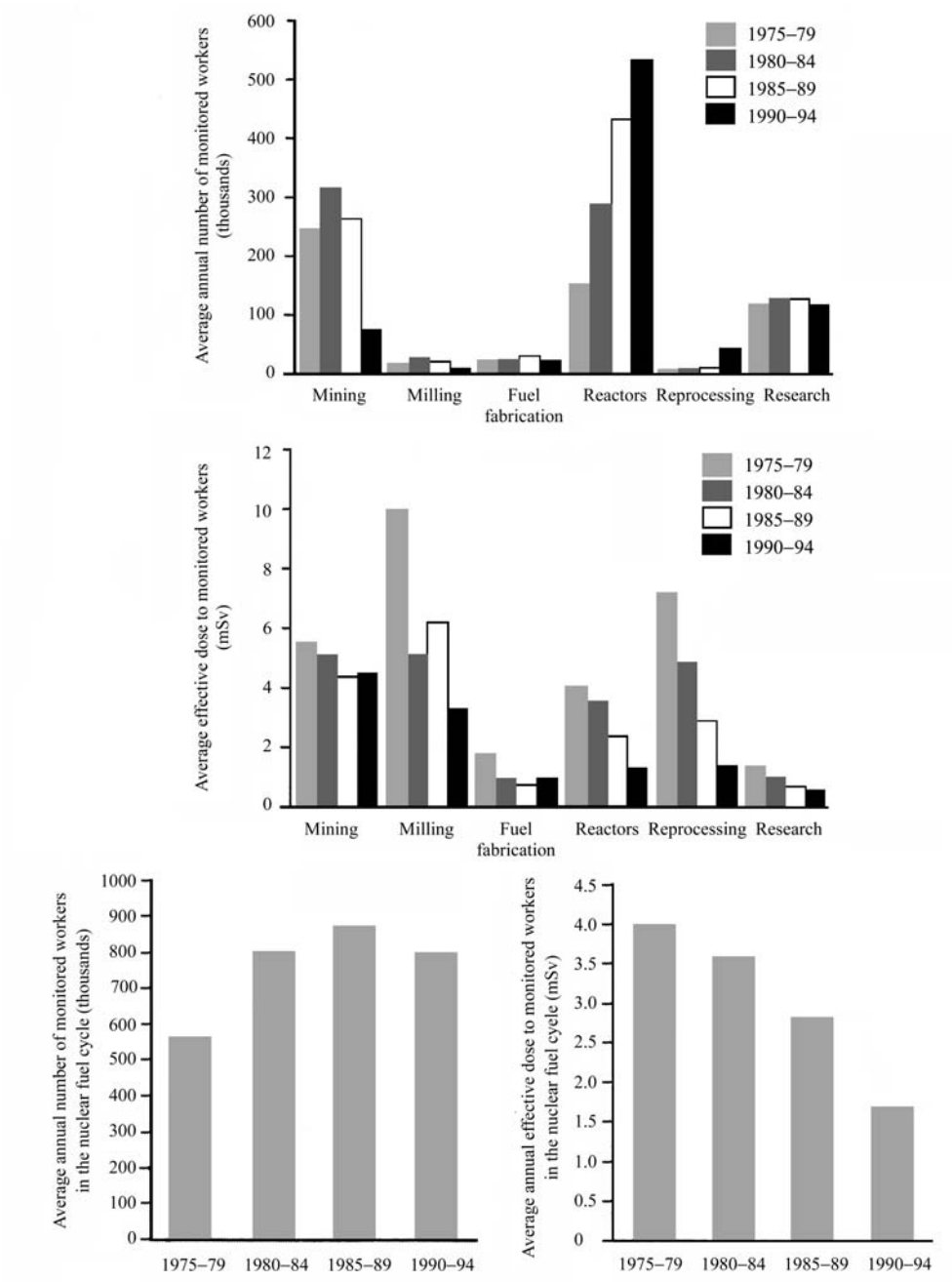
(*h*) *Mineral processing*

There is a substantial worldwide industry in which minerals and ores containing relatively high concentrations of uranium and thorium are mined, milled and processed, either to recover the metals themselves or to obtain the minerals that occur with them. Examples of such ores and minerals are bastneasite (thorium, 5 kBq/kg), monazite (uranium, 6–20 kBq/kg; thorium, 4–7% by weight), phosphate (uranium, 0.1–4 kBq/kg), pyrochlore and columbite (uranium, 50 kBq/kg; thorium, 50 kBq/kg). Exposure to dusts during dry operations in un-enclosed facilities is the major source of internal deposition of the radionuclides. It has been estimated that handling of materials containing activity concentrations of 1–10 kBq/kg could lead to annual effective doses to workers of approximately 1–2 mSv from external and internal exposure (UNSCEAR, 2000).

Mineral sands containing concentrations of thorium up to 8% are mined and processed in Australia, India, Malaysia and South Africa. Minerals recovered in mines in Western Australia are listed in Table 16, the main production activities being directed at ilmenite and zircon; however, monazite (containing 5–7% thorium) is an important component of inhalatory exposure, because it concentrates preferentially in airborne dust. Exposure has been reduced significantly in the Western Australian mineral sands industry: the mean annual dose declined from about 25 mSv (of which 10% was due to internal exposure) to around 6 mSv (15% internal exposure) in the period 1990–94 (UNSCEAR, 2000).

Measurement of ^{220}Rn in the exhaled breath of workers in thorium refineries is often used as an indication of internal contamination with ^{232}Th. These workers inhale the radionuclides ^{232}Th, ^{228}Ra, ^{228}Th, ^{224}Ra and their associated decay products. A certain fraction of ^{220}Rn produced by internally deposited thorium and radium is exhaled in breath. Workers at a thorium-processing plant in Trombay, India, were monitored for internally deposited ^{232}Th by analysis of exhaled air. The results are expressed in Q_{Ra} units, i.e. the equivalent activity (in Bq) of ^{224}Ra measured as ^{220}Rn at the mouth, with

Figure 3. Trends in numbers of monitored workers and doses to workers in the nuclear fuel cycle



From UNSCEAR (2000)

Table 16. Minerals recovered in the mineral sands industry in Western Australia

Mineral	Chemical formula	Per cent of production	Concentration (% by weight)	
			Thorium	Uranium
Ilmenite	$FeOTiO_2$	76	0.005–0.05	0.001–0.003
Monazite	$[Ce, La, Nd, Th]PO_4$	< 1	5–7	0.1–0.5
Rutile	TiO_2	< 5	0.005–0.01	0.001–0.003
Zircon	$ZrSiO_4$	19	0.01–0.025	0.015–0.03
Xenotine	YPO_4	< 1	1.5	0.4

From UNSCEAR (2000)

a lower detection limit of 0.56 Bq. For three categories of workers (100 operators, 55 maintenance workers and 21 supervisors), the Q_{Ra} values were < 0.56 Bq for 33%, 45.5% and 57.2% and 1.11–1.85 Bq for 27%, 18% and 14% of the workers, respectively. Q_{Ra} values of > 1.85 Bq were measured for 19% of the operators, for 1% of the maintenance workers and for none of the supervisors (Mayya *et al.*, 1986).

The Baiyan Obo rare-earth and iron mine is sited in Inner Mongolia in China and is one of the largest rare-earth mines in the world. It has been in operation since 1958. Its ore contains not only iron, rare-earth elements and silica but also 0.04% thorium. In a 14-year follow-up study, the lung burden of thorium was estimated for 638 miners who had inhaled dust, on the basis of measurements of exhaled ^{220}Rn, measured as ^{224}Ra equivalent activity. The average lung burden was estimated to be 1.60 Bq during 1983–84, and the highest value was 11.11 Bq. Of these workers, 585 (91.7%) had lung burdens < 2.22 Bq, while the average lung burden in 143 controls was calculated to be 0.30 Bq (Chen *et al.*, 2000). The airborne concentrations of ^{232}Th were in the range of 0.015–1.41 Bq/m^3, with an average of 0.3 Bq/m^3 (Chen *et al.*, 1993).

(i) *Nuclear weapons production*

The main potential sources of occupational exposure to radionuclides during the development and production of nuclear weapons are ^3H and the two fissile materials plutonium and uranium. Exposure may occur through intake of these materials into the body by inhalation or ingestion (or absorption through the skin in the case of ^3H), and through external irradiation from γ-rays and, to a lesser extent, neutrons. Application of safety measures and containment of the radioactive materials has reduced internal exposure to below external exposure (UNSCEAR, 2000).

Significant internal depositions of plutonium occurred in workers on the Manhattan Project at Los Alamos (USA) in 1944 and 1945. Twenty-six workers were selected in 1951 for long-term follow-up. They were exposed mainly by inhalation, owing to poor working conditions, use of open-faced chemical hoods and inadequate respiratory protection. Tissue contaminated through minor cuts or puncture wounds was excised from seven subjects on the day of the injury. The radioactivity in one of the tissue

samples amounted to 55 Bq, and the other samples contained < 1 Bq. Fifty years after the period of exposure, one subject still had residual activity (310 Bq) around the wound site, but the radioactivity in the excised tissue had not been measured in this case. Internal plutonium deposition was estimated on the basis of measurements of plutonium in urine. The effective doses, calculated as the sum of the annual effective doses through 1994 or the year of death, ranged from 0.1 to 7.2 Sv (mean, 2.08 Sv; median, 1.25 Sv). Direct in-vivo measurements of ^{239}Pu and ^{241}Am in the lungs showed activity over the background level in only one subject, who had 7 Bq of ^{241}Am on his chest count (Voelz *et al.*, 1997).

Kathren *et al.* (1987) reported an interlaboratory study to compare estimates of internally deposited plutonium from urine analysis and from measurements in autopsy tissue of 17 individuals in the Transuranium and Uranium Registry in the USA, after exposure to plutonium. Intake occurred mainly through acute inhalation, skin contamination or through contaminated wounds. Samples of the liver and skeleton, the major reservoir organs for plutonium, were collected at autopsy and analysed radiochemically for ^{239}Pu content. The estimated total body burden of the 17 subjects ranged from 0.01 to 0.77 kBq. Estimates of systemic deposition of plutonium on the basis of analysis of urine were compared with earlier observations (Langham, 1957) in 18 human subjects who were injected experimentally with plutonium citrate. In 16 cases, these estimates were consistently higher (1.5–22-fold) than those from the analysis of tissues collected at autopsy, particularly at lower levels of plutonium. The results suggest that the method used to convert urinary concentrations to body burden results in overestimates of the latter.

Internal exposure to incorporated ^{239}Pu was estimated for 500 workers at the Mayak nuclear enterprise in the southern Urals by analysis of the amount of ^{239}Pu in urine. The absorbed dose to the lung from internal α-radiation was estimated on the basis of the ICRP lung clearance models. The mean ^{239}Pu body burden was 0.01 kBq for 130 workers, 0.34 kBq for 68 workers, 1.2 kBq for 125 workers, 4.2 kBq for 95 workers, 16.5 kBq for 63 workers and 54.2 kBq for 19 workers. The mean absorbed lung dose was 0 Gy for 100 workers, 0.04 Gy for 166 workers, 0.35 Gy for 149 workers, 1.38 Gy for 31 workers, 3.30 Gy for 36 workers and 9.70 Gy for 18 workers. The mean exposure to external γ-radiation, determined from personal film badges, was 0.3 Gy for 230 workers, 1.4 Gy for 117 workers, 2.7 Gy for 144 workers and 5.4 Gy for nine workers (Tokarskaya *et al.*, 1997a).

An updated report on the characteristics of the Mayak workforce (n = 14 416) provided details on the plutonium body burden of 4186 workers (3136 men, 1050 women) on the basis of urinary excretion data. The results are shown in Table 17. In the group with the heaviest exposure (> 3.70 kBq), the average plutonium body burden was 12.91 kBq for men and 29.66 kBq for women. For all workers with a measurable body burden (0.74–3.70 kBq), the overall average values were 2.19 kBq for men and 4.02 kBq for women (Koshurnikova *et al.*, 1999).

Table 17. Distribution of plutonium body burden among workers at the Mayak nuclear plant, Russian Federation

Pu body burden (kBq)	Men	Women
Not detected	413 (13%)	174 (17%)
< 0.74	1492 (48%)	494 (47%)
0.74–1.47	516 (16%)	179 (17%)
1.48–2.95	342 (11%)	94 (9%)
2.96–3.69	64 (2%)	10 (1%)
> 3.70	309 (10%)	99 (9%)

From Koshurnikova *et al*. (1999)

Plutonium intake by workers at the Sellafield plant of British Nuclear Fuels (United Kingdom) was also assessed by monitoring urine samples. Exposure at Sellafield was to so-called 'plutonium α' isotopes (^{239}Pu and some ^{238}Pu and ^{240}Pu) and to ^{241}Pu, which decays by β-particle emission to the α-particle emitter ^{241}Am. Before 1960, the plant mainly produced plutonium for military purposes, and exposure to ^{241}Pu began only after that time, during commercial reprocessing of spent nuclear fuel. The uptake was estimated for about 4600 workers employed at Sellafield for some time between 1950 and 1990. The average annual intake of 'plutonium α' isotopes increased in the first years to 100 Bq per year and then decreased to near background levels in 1965. Intake of ^{241}Pu increased to an average annual value of 250 Bq in 1970, which decreased to 100 Bq in 1990 (Omar *et al.*, 1999).

Operations involving ^{210}Po were conducted at the Mound Facility in the USA from 1944 to 1972. The facility was established to support the Manhattan Project and focused on the separation and chemical preparation of polonium. Urine bioassays to monitor personnel for internal exposure to ^{210}Po were conducted throughout operation of the plant. The data from the urinary analyses were used to compute the estimated doses to the kidney and spleen, that to the kidney being assumed to represent the overall soft-tissue dose. Four dose groups were distinguished: < 10 mSv, 10–100 mSv, 100–1000 mSv and > 1000 mSv. Workers in these four groups had accumulated 40 700, 8000, 6760 and 4300 person–years, respectively, at the Mound facility. The mean effective dose from external radiation (26.5 mSv) was approximately the same for all groups, except for those exposed to > 1000-mSv, who had a mean effective dose of 36.1 mSv from external exposure (Wiggs *et al.*, 1991).

A worker at the Hanford nuclear waste treatment facility in the USA who was injured in 1976 during chemical explosion of an ion-exchange column used for the recovery of ^{241}Am, was monitored until his death in 1987. He was exposed to the radionuclide through inhalation and via wounds on his face and neck. The victim received calcium trisodium diethylene triamine pentaacetate (DTPA) and zinc DTPA as long-term decorporation therapy, with several daily doses during the first month.

A total of 583 g of DTPA was administered between 1976 and 1980, with no apparent toxic effects. Detailed external measurements of internally deposited ^{241}Am were started on the third day after exposure. Lung, liver, bone and the skin of the face and neck were found to have received the highest dose rates. Three days after the accident, the organ burdens were reported to be 26 MBq for skin, 960 kBq for lungs, 480 kBq for bone and 1400 kBq for liver. After 10 years, the values had decreased to 110 kBq for skin, 350 kBq for bone and 19 kBq for liver, with only residual activity (about 2 kBq) in the lungs. The total amount of radionuclide excreted from the body was 41 MBq, about half of which was excreted in the first three days. DTPA therapy appeared to be effective in preventing ^{241}Am deposition in bone and liver; significant re-deposition in the liver occurred upon termination of the DTPA treatment. The only radiation effects were depressed lymphocyte, platelet and neutrophil counts in the peripheral blood and cytogenetic effects in the lymphocytes (Breitenstein & Palmer, 1989).

Data on the exposure of workers in defence activities in the USA related to nuclear weapons are given in Table 18. Combined data for workers in the United Kingdom and the USA in all defence activities (nuclear weapons manufacture and associated activities, operating nuclear vessels and their support facilities) are given in Table 19 (UNSCEAR, 2000).

Table 18. Occupational exposure of workers in defence activities related to nuclear weapons (USA)

Year	Monitored workers (thousands)	Measurably exposed workers (thousands)	Average dose to measurably exposed workers (person–Sv)	Collective effective dose equivalent (person–Sv)		
				External[a]	Internal	Total
1990	108	36.0	0.85	22.3	8.2	30.5
1991	120	31.3	0.82	17.6	8.1	25.7
1992	124	29.4	0.78	15.0	7.9	22.9
1993	127	24.0	0.68	15.3	0.95[b]	16.3
1994	117	25.4	0.65	16.0	0.43	16.4
1995	127	23.6	0.78	18.1	0.31	18.4

From UNSCEAR (2000)
[a] γ-Radiation and neutrons
[b] Considerably less than that in previous years because of changes in calculating internal exposure

(*j*) *Nuclear medicine* (see also section 1.3)

The aim of nuclear medicine is to investigate physiological processes, in most cases by measuring organ function. The use of radionuclide generators, especially those of ^{99m}Tc, involves handling of large amounts of radioactivity. The radionuclides used in organ imaging (e.g. ^{99m}Tc) and in treatment (e.g. ^{131}I) emit penetrating γ-radiation, and

Table 19. Occupational exposure of workers in all defence activities (United Kingdom and USA)

Period[a]	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	104	137	1.3
1980–84	116	82	0.71
1985–89	127	84	0.66
1990–94	139	33	0.24

From UNSCEAR (2000)
[a] Data are annual averages over the periods indicated.

the main exposure of personnel occurs during imaging, i.e. while injecting the patient and positioning the patient and the camera. The internal exposures of personnel are usually much lower than these external exposures. Data on the exposure during nuclear medicine procedures worldwide are given in Table 20. While the number of monitored workers increased during the four five-year intervals studied, the average annual effective dose remained fairly constant at about 1.0 mSv during 1975–90 and decreased to approximately 0.8 mSv during the period 1990–94. Regional variations have been observed, most notably in Latin America and the Indian subcontinent, with a value of about 2.3 mSv (UNSCEAR, 2000).

The increasing number of new radiopharmaceuticals and the increased dose per patient may enhance the radiation burden of radiopharmacy personnel. At present, a medium-sized radiopharmacy may order seven to eight 3-Ci [11.1×10^4 MBq] ^{99}Mo/-^{99m}Tc generators per week and may perform 10–15 elutions per day. The maximum radiation exposure of radiopharmacy personnel occurs during generator elutions, compounding (kit preparation) and unit-dose dispensing. Additional exposure can occur

Table 20. Occupational exposure in nuclear medicine worldwide

Period[a]	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	61	62	1.01
1980–84	81	85	1.04
1985–89	90	85	0.95
1990–94	115	90	0.79

From UNSCEAR (2000)
[a] Data are annual averages over the periods indicated.

during receipt of radioactive packages, transport of prepared doses and storage and disposal of radioactive waste. Doses to the fingers and hands of 140–210 mSv/year have been reported. Such doses can be reduced by a number of techniques, including use of forceps during transfer of radioactive vials and syringes and use of a syringe shield during unit-dose dispensing. Most of the exposures mentioned here are from external radiation. During the preparation of unsealed doses or capsules of radioactive iodine, however, there is potential for a high internal dose from inhalation of volatile solutions (Heller, 1996).

Personnel working with ^{125}I at a Swedish hospital were monitored regularly over 20 years for the ^{125}I content of their thyroids. Four categories were studied: chemists performing ^{125}I-labelling of proteins and hormones, using up to 74 MBq per single labelling; laboratory staff performing mainly radioimmunoassays (approximately 1 kBq ^{125}I per sample); nuclear medicine staff giving intravenous injections of ^{125}I-labelled radiopharmaceuticals (up to 4 MBq per injection) to patients; and administrative staff working in the same departments as the above categories but not handling ^{125}I. Personnel from another department, not working with ^{125}I, were used as a control group. During the period 1977–90, the chemists had a mean ^{125}I activity of 1.5 kBq in their thyroids, giving a yearly absorbed dose of 10 mGy to this organ. A value of 7.4 kBq (53 mGy/year) was recorded on one occasion in one person. The mean activity for laboratory staff was 140 Bq, with an individual value of 4 kBq recorded in this group. For the nuclear medicine staff, an average thyroid activity of 30 Bq ^{125}I was recorded. Control personnel in the same department had received up to 25 Bq, indicating diffuse, low-level contamination with ^{125}I in the work area. Control subjects from elsewhere in the hospital showed no measurable ^{125}I in their thyroids (< 2 Bq). A trend was noted towards decreasing internal contamination with time during the period 1977–90 in spite of constant use of ^{125}I. A further decrease in internal contamination during 1991–1996 was attributed to a decrease in the use of ^{125}I (Jönsson & Mattsson, 1998).

An evaluation was made of the doses received by staff working in a nuclear medicine department and by children who may be in close contact with a parent receiving treatment with radionuclides. Dose rates were measured at distances of 10 cm, 50 cm and 1.0 m from the skin surface at the level of the thyroid, chest and bladder of patients undergoing the following procedures: multiple-gated acquisition scans to determine coronary function with ^{99m}Tc-labelled red blood cells, myocardial perfusion scans with ^{99m}Tc-labelled radiopharmaceuticals, lymphoscintigraphy with colloidal ^{99m}Tc (Re) sulfide, bone scans with ^{99m}Tc-labelled oxidronate, ^{111}In-octreotide scans, ^{111}In-labelled leukocyte studies and cardiac re-injection studies with ^{201}Tl. The maximum dose rates at 10 cm were those in myocardial perfusion (one-day protocol, 391.7 μSv/h; two-day protocol, 121.8 μSv/h) and multiple-gated acquisition scans (167.3 μSv/h). The maximum dose received by an infant in close contact with a parent after a nuclear medicine investigation was estimated to be 1.53 mSv, which arose from close contact (approximately 13 h during a 40-h period following treatment) with a parent who had received a ^{99m}Tc myocardial perfusion on a one-day protocol. It is

unlikely that a nuclear medicine technician would receive an annual dose of more than 6 mSv. Likewise, doses received by the nurses of patients treated with radiopharmaceuticals are probably < 6 mSv/year, except perhaps when they are in regular contact with large numbers of helpless patients (Greaves & Tindale, 1999).

Administration of a radiopharmaceutical to a patient gives rise to a radiation hazard to the patient and, possibly, to critical groups exposed to the patient. Estimation of the effective doses for adult and paediatric patients is limited by uncertainties about the biokinetics and the assumption of a uniform distribution of activity in each organ. In the United Kingdom, the effective doses from most nuclear medicine procedures do not exceed twice the annual dose from natural background radiation. Lack of data on human placental transfer is now the main limitation to estimating fetal doses. Internal exposure to radionuclides may occur during breast-feeding of an infant by a mother who has received treatment with radiopharmaceuticals. To ensure that the effective annual dose to the infant does not exceed 1 mSv, interruption of breast-feeding is recommended in some cases (for 25 h after administration of 80 MBq ^{99m}Tc-pertechnetate) and cessation of breast-feeding in others (treatment with ^{32}P-phosphate, ^{131}I-iodide) (Mountford, 1997).

(*k*) *Luminizing*

Luminizing is one of the oldest industrial uses of ionizing radiation. Employment in the luminizing industry in the USA began around 1913, and about 5000 workers are known to have been employed in this industry between 1915 and 1980. While they were not all involved in dial painting, all were at some risk of occupational exposure to the radiation emitted by the radium present at the worksite. Initially, the luminizing paints were enhanced with ^{226}Ra alone, but later some paints contained a mixture of ^{226}Ra and ^{228}Ra. The greatest risk of internal exposure to radium was that of the early dial painters who used to 'tip' and 'point' their brushes between their lips. Before 1925, the daily intake of radioactive substances is estimated to have been 3–48 μg. 'Tipping' was officially banned in the USA in 1926 (Fry, 1998).

The radium body burdens were measured of two groups of female radium-dial workers in the USA, whose first year of employment was either before 1930 (n = 693) or between 1930 and 1949 (n = 561). Measurements were performed by γ-ray spectroscopy of ^{228}Ra *in vivo* and by analysis of radon in exhaled air. The proportions of ^{226}Ra and ^{228}Ra in the paint used in the dial industry before 1930 varied over time and among companies, some factories often using paints very rich in either ^{228}Ra or ^{226}Ra. The estimated total radium intakes of the group employed before 1930 were < 0.5 μCi [about 20 kBq] for 29% of the women, 0.5–4.9 μCi [about 20–200 kBq] for another 29%, 5–50 μCi [about 200–2000 kBq] for 26% and > 50 μCi for 16%. During 1930–49, the radium body burdens were approximately 100-fold lower than before 1930, and ^{228}Ra was not in use during this period. Only 1% of the 561 workers in this group had estimated intakes above 4.9 μCi; 32% had intakes of 0.5–4.9 μCi, and 67% had estimated intakes of < 0.5 μCi (Stebbings *et al.*, 1984).

The United Kingdom radium luminizer survey covered 1110 women who had worked as luminizers between 1939 and the late 1950s. Luminizing was not widespread in the United Kingdom until the Second World War, when there was a demand for luminous aircraft instruments. On the basis of the experience in the USA during the 1920s and 1930s, the intake of radium during painting was more strictly controlled, leading to much lower exposure levels. The body burdens of the 459 women monitored were < 0.01 μCi [370 Bq] ^{226}Ra for 197, 0.01–0.03 μCi [370–1110 Bq] for 202 and > 0.03 μCi for 60 women. From a retention model of alkaline earths in the body, it was estimated that the mean systemic intake of these groups had been 22, 3 and 0.4 μCi [814, 111 and 15 kBq], respectively. If it is assumed that the systemic intake was 0.02 Sv/μCi, the committed doses to the red bone marrow would have been 440, 60 and 8 mSv (Baverstock & Papworth, 1985).

The decay of ^{226}Ra deposited in the bone of workers in the luminizing industry gives rise to formation of ^{210}Pb and ^{210}Po *in situ*, and these radionuclides may be transferred away from the site of deposition of the radium. In one case, the bone of a former dial painter contained ^{226}Ra at about 4000 pCi/kg [148 Bq/kg] and ^{210}Po at 1500 pCi/kg [56 Bq/kg], the ratio of bone:soft-tissue concentration being much higher for ^{210}Po than after direct intake of this radionuclide. The highest concentration of polonium in any tissue in the radium-dial painters was found in hair, one sample containing 25 μCi/kg [925 kBq/kg] (Hill, 1965).

With time, there has been a shift in the luminizing industry from use of radium to use of ^{3}H and, to a lesser extent, ^{147}Pm. The number of workers involved during 1975–94 was small (< 1000). The number of workers monitored during 1990–94 was about 80, and the average annual effective dose was 0.4 mSv (UNSCEAR, 2000).

(*l*) *Radioisotope production and distribution*

Occupational exposure during the production and distribution of radioisotopes for a wide variety of industrial and medical purposes stems mainly from external irradiation. Internal exposure has generally not been included in the exposure data, but when this was reported, e.g. for the United Kingdom from 1985 and for Finland from 1987 onwards, the contribution of internal exposure to the total dose was estimated to be a few per cent. Worldwide, the estimated total number of workers involved in radioisotope production and distribution increased from 57 000 to 88 000 in the period 1975–89 and decreased to 24 000 in 1990–94. This decrease probably reflects rationalization of production and concentration of activities by multinational companies in a limited number of places. The estimated worldwide annual collective effective dose decreased from more than 130 person-Sv in 1975–79 to 47 person–Sv in 1990–94, and the annual effective dose per monitored worker decreased from 2.3 to 1.9 mSv over 1975–94 (UNSCEAR, 2000).

(*m*) *Research centres*

Research workers in universities, polytechnics and research institutes may use radioactive labels, e.g. ^{3}H, ^{14}C, ^{32}P, ^{35}S, ^{125}I, and sealed sources containing ^{60}Co or ^{137}Cs. Exposure data are given in Table 21. The apparent doubling of the number of workers monitored in the period 1990–94 may be an overestimate, which can be attributed to the method used, i.e. extrapolation within regions on the basis of gross domestic product. The total annual collective effective dose decreased from 74 to 22 person–Sv in the period 1975–89 and increased to 33 person–Sv over the years 1990–94. The annual average effective dose decreased from 0.55 to 0.11 mSv during 1975–94 (UNSCEAR, 2000).

Table 21. Occupational exposure in educational establishments worldwide

Period[a]	Monitored workers (thousands)	Total annual collective effective dose (person–Sv)	Average annual effective dose (mSv)
1975–79	140	74	0.55
1980–84	180	43	0.24
1985–89	160	22	0.14
1990–94	310	33	0.11

From UNSCEAR (2000)
[a] Data are annual averages over the periods indicated.

1.3 Medical use of radionuclides

The use of radionuclide preparations for medical purposes, referred to as ‘radiopharmaceuticals’, is widely practised throughout the world; ‘nuclear medicine’ is the term used to describe medical diagnostic and therapeutic techniques based on the use of radiopharmaceuticals. Radiopharmaceuticals have been used for therapeutic purposes for more than 60 years, but external beam therapy or encapsulated (sealed) source therapy are by far the most common therapeutic radiation modalities (Carlsson, 1995). This section does not deal with the use of radiopharmaceuticals for analysis of biological specimens, such as blood and urine, in radioimmunoassay procedures but focuses on administration of radiopharmaceuticals for diagnostic and therapeutic reasons in humans.

1.3.1 *Diagnostic use*

(*a*) *Procedures*

Decreased or absent function of an organ, due for instance to the presence of a tumour, can be inferred from a reduced concentration of an appropriate radiopharmaceutical. Tracer tests, scans, renograms, metabolic and haematological examinations are, however, somewhat different from diagnostic radiology, as the results provide information on the physiological process during the short period between administration of the radionuclide and imaging. After administration to a patient, the distribution and localization in the tissue is determined by the pharmaceutical preparation used. Nuclear medical examinations thus give both functional and quantitative information.

One of the characteristics of nuclear medical examinations is that pathological processes can be diagnosed before other diagnostic methods can show morphological alterations; metastases in the skeleton can often be shown months before the first clinical symptom or even before detection at conventional X-ray examinations. The principles of some of the most common nuclear medical examinations are shown in Table 22.

Table 22. Clinical use of common nuclear examinations

Examination	Clinical use
Bone scintigram	Osteoblast activity
Lung scintigram	
Perfusion	Capillary perfusion
Ventilation	Distribution of ventilation
Renogram	Glomerular filtration rate, tubular secretion
Thyroid scintigram	Iodine metabolism
Myocardial scintigram	Myocardial metabolism, circulation and perfusion

Technical developments, computer-assisted imaging techniques and better cameras have improved the quality of diagnostic procedures. In addition to the conventional two-dimension imaging, techniques have been developed to allow emission tomography which shows internal structures in cross-sectional ‘slices’ of a patient. The commonest technique is single-photon emission computed tomography; a more specialized procedure is positron emission tomography, based on simultaneous detection of pairs of photons arising from positron annihilation (Meyer *et al*., 1995; Rigo *et al*., 1996; Schiepers & Hoh, 1998; Ferrand *et al*., 1999; UNSCEAR, 2000).

The latest report from UNSCEAR (2000) included a survey of the use of diagnostic nuclear medicine worldwide. As can be seen from Table 23, thyroid examinations (scans and uptake tests) constituted approximately 28% of all procedures in the countries listed, followed closely by bone scans (26%) and cardiovascular examinations (15%). Some of the commoner examination procedures are described briefly below.

Table 23. Average annual distribution (%) by type of diagnostic nuclear medicine procedure and country, 1991–96

Country or region[a]	Bone	Cardio-vascular	Lung		Thyroid		Kidney	Liver/spleen	Brain
			Perfusion	Ventilation	Scan	Uptake			
Argentina	30	27	2.9	2.3	16	11	7.4	1.2	1.9
Belarus	48	–	–	–	2.4	0.8	35	0.4	0.4
Bulgaria	2.1	2.0	1.4	0.4	38	45	6.8	1.7	1.6
Canada	34	47	0.3	1.5	4.3	4.6	2.5	0.9	2.4
China, Taiwan Province	23	15	2.1	–	4.7	5.3	4.4	20	9.8
Croatia	22	11	2.8	0.3	23	1.5	27	2.4	5.3
Cyprus	27	27	1.8	0	21	0.02	16	0.3	0
Czech Republic	18	8.6	9.5	1.4	9.2	3.5	29	4.1	7.7
Denmark	19	8.5	5.9	3.6	13	2.0	23	0.1	2.2
Ecuador	32	7.2	3.7	1.9	27	21	3.6	2.7	0.2
Finland	39	13	12	2.2	1.7	0.9	17	0.1	2.8
Germany	26	8.3	7.6	–	50	–	4.7	0.1	1.4
Hungary	26	6.5	6.8	0.6	27	4.4	17	2.5	2.2
Ireland	45	5.3	11	2.5	0.8	1.7	24	0.4	0.2
Italy	33	14	4	0.6	23	2.3	12	3.2	3.2
Japan	24	7.0	3.9	–	8.1	–	6.0	5.3	11
Kuwait	7.1	20	2.1	–	31	16	7.7	0.6	0.4
Lithuania	3.2	0.1	0.2	0	16	16	10	1.3	0.01
Netherlands	39	20	7.0	7.3	5.3	3.1	7.6	0.6	1.6
New Zealand	49	7.3	9.0	6.9	7.9	0.3	10	1.0	3.0
Panama	5.2	5.7	5.5	6.5	50	11	7.1	5.0	3.8
Qatar	25	20	3.5	–	12	–	29	1.4	0
Romania	12	–	1.0	–	27	20	9.4	26	3.5

Table 23 (contd)

Country or region[a]	Bone	Cardio-vascular	Lung		Thyroid		Kidney	Liver/spleen	Brain
			Perfusion	Ventilation	Scan	Uptake			
Slovakia	29	2.6	16	–	27	0.05	9.4	6.6	0.5
Slovenia	18	12	7.3	4.0	23	3.4	13	1.5	4.3
Sweden	28	7.9	11	4.4	9.0	3.8	3.1	0.6	0.7
Switzerland	43	5.6	14	6.4	15	–	4.2	0.5	1.8
United Arab Emirates	27	15	2.3	0.3	13	13	19	1.3	0.6
USA	24	13	16	–	–	–	3.2	22	11
Average	26	15	10	2.0	23	5.3	5.0	12	7.3

From UNSCEAR (2000). Since not all procedures are listed, the rows do not add up to 100%.

[a] Health-care level I countries or regions (see Table 25)

(i) *Thyroid examinations*

The possibility of comparing thyroid tissue function (iodine metabolism, hormone synthesis) with an evaluation of the thyroid gland by radionuclide scanning was a milestone for nuclear medicine and endocrinology. Thyroid scans with ^{131}I, ^{123}I or ^{99m}Tc to identify 'hot' and 'cold' nodules are widely used. After clinical examination of the thyroid gland, patients may be referred for a thyroid scan, fine-needle biopsy or ultrasound examination. Ultrasound examination of the thyroid gland has reduced the indications for thyroid scans, but the latter method is still widely used to measure the function of the thyroid gland and to reveal the morphological pattern. Function can also be determined from thyroid hormone measurements, but actual iodine uptakes are used to guide future therapeutic administration of radioiodine. Thyroid scans reveal activity in different parts of the gland, the size and number of pathological findings and atypical thyroid tissue (Carlsson, 1995; Nusynowitz, 1999).

(ii) *Bone examinations*

The second most common use of radiopharmaceuticals for examination is bone scanning for staging of tumours and diagnosis of infectious or inflammatory processes in the skeleton. Skeletal scintigraphy provides information on metabolic changes produced by modifications of bone vascularity and osteoblastic activity. Strontium and fluorine were used for skeletal scintigraphy, but they have been totally replaced by compounds containing ^{99m}Tc, reflecting the ideal properties of this radionuclide for clinical use (Feith *et al.*, 1976; O'Mara, 1976; Carlsson, 1995).

(iii) *Cardiovascular examinations*

For many years, studies of vascular transit and cardiac function were the only nuclear cardiovascular examinations known. Perfusion imaging developed more slowly. In the 1970s, a major breakthrough came with the visualization of myocardial necrosis. The development of single-photon emission computed tomography and positron emission tomography introduced a vast number of perfusion tests, many with the aim of detecting coronary artery disease (Saha *et al.*, 1992; Carlsson, 1995; Lee *et al.*, 2000).

(iv) *Pulmonary perfusion and ventilation tests*

Pulmonary embolism is a potentially fatal complication of deep-vein thromboses, but the clinical presentation of pulmonary embolism is non-specific and additional evaluation with imaging studies is essential for obtaining an accurate diagnosis. Scintigraphic examination of the lungs has been designed to demonstrate patterns of ventilation and perfusion. Pathophysiological conditions are commonly associated with scintigraphically detectable perturbations. Perfusion scintigraphy of the lung is accomplished by microembolization of radionuclide-labelled particles in the pulmonary arterial circulation. Albumin-labelled ^{99m}Tc is commonly used. For ventilation imaging, ^{133}Xe gas is inhaled by the patient. During wash-out, rapid clearance of activity from the lungs is seen, but areas of abnormal ventilation become apparent as hot spots (Taplin, 1979; Carlsson, 1995).

(*b*) *Doses*

Most radionuclides used in the field of nuclear medicine are produced by cyclotron bombardment, reactor irradiation, fission products or generators of secondary decay products from long-lived parent radionuclides. Only a few of the approximately 1700 known radioactive isotopes are suitable for medical purposes, as certain physical and chemical characteristics are required. Low photon energies give images with inferior resolution, whereas high photon energies result in an unacceptably high absorption of energy in the patient. A radionuclide with a long half-life gives a high absorbed dose to the patient, and a short half-life could result in a low concentration of the radionuclide in the organ under examination. The carrier molecule used for transporting the radionuclide must be stable enough to secure measurements characteristic of the organ or the process that is being examined. Electrons and positrons are not particularly helpful in the diagnostic setting, contrary to nuclear medical therapeutic techniques in which localized absorption of ionizing radiation is important (Carlsson, 1995; UNSCEAR, 2000).

Most of the radionuclides used in diagnostic and therapeutic treatments are listed in Table 24. Those most commonly used are ^{99m}Tc and ^{131}I. The half-life of ^{99m}Tc is 6 h, and it emits photons with an energy of 140 keV. Radioiodines have been used for nearly 60 years for diagnostic and therapeutic purposes: ^{131}I (half-life, eight days) gives a high photon energy and is fairly cheap; ^{123}I emits photons with an energy of 159 keV and is therefore well suited for diagnostic purposes; however, it is quite expensive and has a half-life of 13 h which makes it difficult to transport over long distances.

The estimated annual number of nuclear medical examinations in the world in 1985–90 was 24 million, corresponding to a frequency of 4.5 per 1000 individuals. The most recent survey indicated 32.5 million in 1991–96, corresponding to 5.6 examinations per 1000 individuals (UNSCEAR, 1993, 2000). The distribution of examinations is, however, highly dependent on the health care level, and 93% of all examination were confined to the western world (Table 25). When data from different parts of the world are compared, it should be remembered that the averages are often based on small sample sizes, particularly for health care levels III–IV. The contributions to frequency and annual collective dose of the commonest examination procedures are given in Table 26. The much higher collective dose from thyroid examinations in less-developed countries is a reflection of the use of the cheaper and more stable ^{131}I instead of ^{99m}Tc. The estimated annual collective effective dose from nuclear medical examinations in 1991–96 to the world population was estimated to be 150 000 person–Sv (Table 27). Many of the patients exposed are near the end of their lives, and the doses are not distributed evenly in the population; thus, these doses should not be used for assessing adverse effects (UNSCEAR, 2000).

Table 24. Radionuclides used for diagnostic and therapeutic purposes

Radionuclide	γ-ray energy (keV)	Half-life	Characteristics of radiation
^{3}H	–	12.3 years	'Pure' β
^{11}C	511	20.3 min	Positron
^{14}C	–	5730 years	'Pure' β
^{13}N	511	10.0 min	Positron
^{15}O	511	124 sec	Positron
^{18}F	511	109.8 min	Positron
^{32}P	–	14.3 days	'Pure β'
^{51}Cr	322	27.8 days	
^{57}Co	122	271 days	
	136		
^{67}Ga	92	3.2 days	
	184		
	296		
	388		
^{75}Se	121	120 days	
	136		
	265		
	280		
	401		
^{89}Sr	–	51 days	'Pure β'
^{90}Y	–	64 h	'Pure β'
^{99m}Tc	140	6.02 h	Most widely used radionuclide
^{111}In	173	2.8 days	
	247		
^{123}I	159	13 h	
^{131}I	284	8.05 days	High-energy photons, high proportion of β-radiation
	364		
	637		
^{133}Xe	81	5.3 days	β^-
^{153}Sm	70	47 h	β^-, Auger-e^-
	103		
^{165}Dy	95	2.3 h	β^-
^{186}Re	137	91 h	β^-
^{198}Au	412	2.7 days	β^-
^{201}Tl	135	73 h	
	167		
^{211}At	–	7.2 h	α

Table 25. Average temporal trends in annual frequency of diagnostic nuclear medicine procedures per 1000 individuals

Health care level[a]	1970–79	1980–84	1985–90	1991–96
I	11	6.9	16	19
II	0.9	0.1	0.5	1.1
III	0.25	0.25	0.30	0.28
IV	–	–	–	0.02

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

Table 26. Contribution to frequency and collective dose of various types of diagnostic nuclear medicine procedures assumed for 1991–96

Procedure	Health care level[a], contribution (%)				
	I	II	III	IV	All
Contribution to total annual frequency					
Bone	24	21	19	8	24
Cardiovascular	14	15	6	0.1	14
Lung perfusion	10	2	2	0.4	9
Lung ventilation	2	1	0.1	0.1	2
Thyroid scan	22	27	59	19	22
Thyroid uptake	5	3	–	42	5
Kidney	5	14	7	13	6
Liver/spleen	11	8	2	1	11
Brain	7	4	4	16	7
All	100	100	100	100	100
Contribution to total annual collective dose					
Bone	25	14	4	2	23
Cardiovascular	27	18	4	0.1	25
Lung perfusion	3	0.6	0.3	< 0.1	3
Lung ventilation	0.4	0.1	< 0.1	< 0.1	0.4
Thyroid scan	10	40	89	28	17
Thyroid uptake	17	10	–	62	16
Kidney	2	6	1	2	2
Liver/spleen	4	2	0.2	0.1	4
Brain	10	4	1	5	8
All	100	100	100	100	100

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

Table 27. Estimated doses to the world population from diagnostic nuclear medical procedures, 1991–96

Health care level[a]	Population (million)	Annual per-capita effective dose (mSv)	Annual collective effective dose (person–Sv)
I	1 530	0.081	123 000
II	3 070	0.008	23 000
III	640	0.006	3 500
IV	565	0.0003	200
World	5 800	0.026	150 000

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

1.3.2 *Therapeutic use*

External beam radiotherapy (teletherapy) or encapsulated source therapy (brachytherapy) are far commoner modalities of radiotherapy than radiopharmaceuticals. Nuclear medical treatment plays a small but important role in the management of patients with cancer, mainly in the palliative setting, and for a few benign conditions such as hyperthyroidism and arthritis. For several benign disorders, radiopharmaceuticals are an alternative to other treatments, and for the treatment of malignant disorders they combine the selectivity of brachytherapy with that of systemic activity (Hoefnagel, 1991; UNSCEAR, 2000).

Radionuclides should be readily available, penetrate a few millimetres, deliver a sufficiently high dose rate and be cheap. Radionuclides that emit medium-energy β-radiation are currently used (Johansson *et al*., 1984; Volkert *et al*., 1991; Johansson *et al*., 1992; Stabin *et al*., 1999). Small ions or molecules that follow physiological pathways, such as [^{131}I]sodium iodide for the treatment of thyroid carcinoma, [^{32}P]orthophosphate for polycythaemia vera and [^{89}Sr]strontium chloride for skeletal metastases, are in common use (UNSCEAR, 2000). Efficient biological targetting can also be implemented with monoclonal antibodies, but such techniques are not yet used in routine clinical practice (McDevitt *et al*., 1998). Some of the current treatment modalities are listed in Table 28, but it should be emphasized that only the first four examples can be considered to be established treatments (UNSCEAR, 2000).

In a recent survey of nuclear medical practice in 17 European countries, 71% of the therapeutic actions in patients with malignant disorders concerned patients with thyroid cancer, 20% concerned palliation of skeletal metastases, 5% concerned treatment for polycythaemia vera and 2% concerned neural crest tumours (Hoefnagel *et al.*, 1999).

The annual numbers of radiopharmaceutical therapeutic interventions in relation to health care level are listed in Table 29 (UNSCEAR, 2000). The calculations are

Table 28. Therapeutically used radionuclides

Radionuclide	Clinical use
^{131}I	Differentiated thyroid carcinomas
^{32}P	Polycythaemia vera
^{89}Sr	Skeletal metastases
^{131}I	Neural crest tumours
^{153}Sm	Skeletal metastases
^{186}Re	Skeletal metastases
^{32}P	Intracavitary tumours
^{90}Y	Hepatic and various other tumours
^{114m}In	Lymphoma
^{131}I	Hepatic and various other tumours

From UNSCEAR (2000)

Table 29. Annual number of radiopharmaceutical therapeutic treatments per 1000 individuals in 1991–96 worldwide

Disease	Health care level[a]					% contribution to world total
	I	II	III	IV	All	
Thyroid malignancy	0.035	0.01	0.003	0.00001	0.015	23
Hyperthyroidism	0.11	0.019	0.017	0.00035	0.042	65
Polycythaemia vera	0.003	0.0001	0	0	0.001	1
Bone metastases	0.005	0.002	0.001	0	0.002	4
Synovitis	0.007	0.0001	0	0	0.002	3
All treatments	0.17	0.036	0.021	0.0004	0.065	96[b]

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

[b] Since not all treatments are listed, the % contribution does not add up to 100%.

based on the distribution of treatments in different countries and average total frequencies for each health care level. As can be seen, the treatment of thyroid disorders comprises nearly 90% of the procedures in the world, and this fraction is even higher in less-developed countries. Almost 75% of the treatments worldwide are performed in economically developed countries, and use of these procedures is more than 400 times commoner in countries with health care level I than in those with level IV. In all countries, therapy for hyperthyroidism predominates (UNSCEAR, 2000).

Table 30 lists the three commonest therapies and the mean administered activity in relation to health care level. In general, the amount of radioiodine administered for hyperthyroidism is one-tenth that for the treatment of thyroid cancer (UNSCEAR,

Table 30. Average activities administered (MBq) in therapeutic treatments with radiopharmaceuticals in relation to health care level, 1991–96, worldwide

Health care level[a]	Thyroid malignancy (^{131}I)	Hyperthyroidism (^{131}I)	Polycythaemia vera (^{32}P)
I	4760	415	170
II	3510	340	148
III	3700	300	–
IV	3500	220	–

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

2000). The annual number of nuclear medical treatments was approximately 400 000 in 1991–96 (Table 31), and, as previously stated, most were performed in the western part of the world. The data in Tables 29–31 indicate that the approximate amount of ^{131}I used annually for the treatment of thyroid cancer is only about 3.7×10^8 MBq, while the corresponding figure for hyperthyroidism is about 0.93×10^8 MBq.

A number of national surveys have been undertaken on nuclear medicine practice and are summarized in the latest report of UNSCEAR (2000). The role of therapeutic nuclear medicine is expanding in developed countries, particularly in oncological practice and not only for differentiated thyroid cancer (Kobayashi *et al.*, 1981; Daghighian *et al*., 1995). The full potential of nuclear medicine will not be realized until target-specific carrier molecules, such as antibodies, have been developed; work is in progress to identify DNA-targetting molecules to enhance cytotoxicity.

Table 31. Estimated annual numbers of therapeutic treatments with radiopharmaceuticals in the world, 1991–96

Health care level[a]	Population (millions)	Annual number of treatments (millions)	Annual number of treatments per 1000 individuals
I	1530	0.3	0.2
II	3070	0.1	0.04
III	640	0.01	0.02
IV	565	0.0002	0.0004
All	5800	0.4	0.065

From UNSCEAR (2000)

[a] Health care level I, one physician per 1000; II, one physician per 1000–3000; III, one physician per 3000–10 000; IV, < one physician per 10 000 persons

1.4 Other applications of radionuclides that could lead to exposure

A variety of consumer products and miscellaneous sources of ionizing radiation result in low levels of exposure of human populations. The major source of exposure to radionuclides is from tobacco products. Examples of other sources of internal exposure are gas lantern mantles, transport of radiopharmaceuticals, smoke detectors and tungsten welding rods (Table 32).

Moeller (1996) categorized consumer products in the USA into five groups, depending on the number of people exposed and the associated dose equivalent. Group 1 includes large numbers of people exposed to sources such as building materials, domestic water supplies, mining and agricultural products, combustible fuel (including natural gas heaters and cooking ranges) and road construction materials. Group 2 includes people for whom exposure is limited to parts of the body. The sources include radioluminous products (including watch dials), gas and aerosol (smoke) detectors, spark gap irradiators and thorium products (including fluorescent lamp starters and gas mantles). Group 3 includes a few people exposed to thorium products such as tungsten welding rods and airport luggage scanning systems. None of these sources represents a significant component of the overall collective dose to populations. Group 4 consists of pilots transporting radiopharmaceuticals, and group 5, people exposed to radiation from cigarettes.

Exposure to these consumer products, used in everyday life, can occur in the home, at work and in the general environment. Some of the best-known sources of exposure are luminous-dial watches, smoke detectors, uranium in ceramic colour glazes, thorium in optical glass, potassium in food products, uranium in construction materials and radon in domestic water supplies. While human exposures from consumer products are small in comparison with those from other sources (such as medical radiation or natural background), large numbers of people are nevertheless exposed (National Council on Radiation Protection and Measurements, 1987a; Schmitt-Hannig *et al.*, 1995; Moeller, 1996).

1.4.1 *Products in the home*

Uranium, radium, thorium and potassium can occur naturally in a variety of building materials, thus increasing population exposure to ionizing radiation. Natural gas used to heat homes and for cooking is a source of airborne radon and its decay products. Similarly, water supplies can result in an increased intake of naturally occurring radioactive materials, as radon can be released during showering, washing clothes, washing dishes and flushing toilets. Naturally occurring indoor radon is discussed in section 1.1.4. The commonest types of smoke detectors contain ^{241}Am, a man-made radioactive material, which ionizes the air between two electrodes through α-particle emissions. When smoke passes through the electrode gap, the resistance is decreased or increased and the associated change is amplified to signal an alarm. The exposure of any individual to

Table 32. Examples of sources and doses of radiation from consumer products in the USA

Product	No. of people exposed	Average annual effective dose (μSv)	Annual collective dose (person–Sv per year)
Group 1 — large numbers of people receiving relatively large doses			
Building materials	125 000 000	70	8750
Groundwater supplies	100 000 000	10–100	5000
Agricultural fertilizers	200 000 000	5–50	2000
Natural gas indoors	135 000 000	4–l8	800
Road construction materials	5 000 000	40	200
Group 2 — large numbers of people receiving relatively small doses or limited to a small portion of the body			
Television receivers	250 000 000	≤ 5	≤ 1250
Video display terminals	50 000 000	≤ 10	≤ 500
Luminous watches	50 000 000	0.1–3	150
Gas lantern mantles	50 000 000	2	100
Transport of radiopharmaceuticals (passengers)	14 000 000	2–3	30
Dental products	45 000 000	0.7	30
Smoke detectors	100 000 000	0.08	10
Airport luggage scanning systems (passengers)	50 000 000	< 0.01	0.5[a]
Group 3 — small numbers of people receiving relatively large doses			
Tungsten welding rods	300 000	160	50
Airport luggage scanning systems (operators)	10 000	1000–2000	15
Thickness gauges	15 000	< 1000	< 15
Transport of radiopharmaceuticals (flight attendants)	30 000	35	
Static eliminators	50 000	3–4	0.2
Group 4 — small numbers of people receiving relatively small doses			
Transport of radiopharmaceuticals (pilots)	15 000	0.7	0.01
Group 5 — tobacco products			
Cigarettes	50 000 000	13 000	650 000

From the National Council on Radiation Protection and Measurements (1987a), modified by Moeller (1996)

[a] Based on an annual average of 10 trips for each member of the public in the USA who flies

these products is negligible. Similarly, certain types of porcelain and bathroom tiles in homes are glazed with uranium; ingestion of radioactive materials by people using such porcelain appears to be minimal, but in some instances their use has been discouraged (National Council on Radiation Protection and Measurements, 1987a; Schmitt-Hannig *et al.*, 1995; Moeller, 1996).

1.4.2 *Personal products*

Radioactive materials are found in a number of food products, luminous-dial watches and clocks, eye pieces, false teeth, jewellery and cigarettes, as discussed previously. ^{40}K accompanies nonradioactive potassium in bananas and other vegetables and fruits, and certain agricultural food products contain small amounts of ^{226}Ra, ^{230}Th and ^{238}U owing to use of these radionuclides in the production of phosphorus-containing fertilizers. In the past, ^{226}Ra was incorporated into numerous timepieces, compasses and other products such as military gauges and instrument dials. Most watches containing ^{226}Ra are no longer in use and were last produced in the USA around 1968: It has been estimated that perhaps one million timepieces containing ^{3}H and ^{147}Pm are now sold yearly in the USA. While the dose rates from ^{226}Ra-containing wrist watches might be as high as 3 mSv/year, the watches containing low-energy ^{3}H and ^{147}Pm result in extremely low doses (National Council on Radiation Protection and Measurements, 1987a; Schmitt-Hannig *et al.*, 1995; Moeller, 1996; IARC, 2000).

Eye glasses and eye pieces in optical instruments contain small amounts of uranium and thorium. In the past, slightly increased dose rates to the germinal cells of the cornea of the eye were reported, and manufacturers subsequently reduced the content of radioactive material in optical glass. Eye glasses that were pink or had rose-coloured lenses incorporated the largest amounts of radioactive materials. In the past, uranium salts were added to false teeth to give them a ‘natural’ colour, which could result in increased exposure of the gums of people using such prostheses, but uranium is no longer incorporated into false teeth. Uranium has been also used as a glaze in jewellery, and some topaz gemstones are slightly radioactive after irradiation with neutrons in order to enhance their blue colour. Such jewellery is regulated. Gas lanterns with incandescent gas mantles, mainly used by campers, can result in exposure to radiation from the small amounts of thorium oxide; however, mantles with no radioactive substances are now available (National Council for Radiation Protection and Measurements, 1987a; Schmitt-Hannig *et al.*, 1995).

1.4.3 *Products in the workplace*

Radionuclides can also be used in static eliminators (e.g. ^{210}Po), thickness gauges, thoriated tungsten welding rods, fluorescent lamps (e.g. ^{85}Kr), spark gaps (e.g. ^{60}Co) and other devices. In general, the dose rates are relatively low. Static eliminators are used in industry to reduce the electric charge build-up on materials like printing

presses, photocopying machines, phonograph records and photographic film (National Council for Radiation Protection and Measurements, 1987a; Schmitt-Hannig *et al.*, 1995; Moeller, 1996).

1.4.4 *Exposure to radionuclides in cigarette smoke*

In the 1960s, ^{210}Pb and ^{210}Po were measured in tobacco, in cigarette smoke and in smokers' lungs (Radford & Hunt, 1964; Hill, 1965; Holtzman & Ilcewicz, 1966; Blanchard, 1967). Several radionuclides have been identified in tobacco smoke, such as radium and thorium, but over 99% of the α-activity results from ^{210}Po (Cohen *et al.*, 1980). Radionuclides accumulate in tobacco plants by either root uptake from soils and fertilizers (Tso *et al.*, 1966) or surface deposition of the nuclides through rainfall (Francis *et al.*, 1968).

1.4.5 *Miscellaneous products and sources*

Spacecraft powered by nuclear sources could result in population exposures if a craft were destroyed during launch or re-entry into the earth's atmosphere. While this has occurred in the past, the associated exposure of individuals in the population was low, partly because the radionuclides were widely dispersed on burn-up of the fuel core in the atmosphere (UNSCEAR, 1993).

Thus, while populations are exposed to radionuclides from a variety of consumer products, the associated doses are usually very small, particularly in comparison with the radiation received annually from natural or medical sources; the possible exception is radioactive polonium in cigarette smoke (Table 32).