EVALUATION OF THYROID DOSES FROM RADIOIODINE RELEASES

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INTRODUCTION

Iodine is a volatile element which is very mobile in the environment. Iodine enters the metabolism of living organisms and is selectively taken up and concentrated in the thyroid gland; it plays a major role in the synthesis of the thyroid hormone and is partly secreted in milk.

There are at least 25 iodine isotopes with mass numbers ranging from 117 to 141. All except ^{127}I are radioactive. Omitting the very short-lived ^{140}I and ^{141}I, thirteen isotopes are produced by fission: ^{127}I (stable), ^{128}I (25 min), ^{129}I (1.57 x 10^7 a), ^{130}I (12.4 h), ^{131}I (8.06 d), ^{132}I (2.3 h), ^{133}I (21 h), ^{134}I (52.8 min), ^{135}I (6.7 h), ^{136}I (83 s), ^{137}I (23 s), ^{138}I (5.9 s), and ^{139}I (2 s). From the point of view of environmental contamination and resulting doses to man, the most important isotope of iodine is ^{131}I, with minor roles occasionally played by the short-lived ^{132}I and ^{133}I, and by the long-lived ^{129}I. Because of its half-life of approximately 8 d, ^{131}I is a short-term problem: within two months after its environmental release, ^{131}I will have decayed to insignificant levels.

Iodine-131 is mainly found in the environment as a result of nuclear explosions and of airborne releases from nuclear reactors and fuel reprocessing plants. Thyroid doses are, in most cases, mainly due to the consumption of ^{131}I-contaminated fresh cows' milk, with minor contributions resulting essentially from the consumption of leafy vegetables and other foodstuffs with short shelf life, and from inhalation. Because of the smaller mass of their thyroid gland, children receive higher doses than adults for a given intake of ^{131}I. Atmospheric releases of, and thyroid doses from, ^{131}I from the nuclear industry are relatively well documented (1-2):

- Testing of nuclear weapons in the atmosphere, which took place essentially in the early 1960s, caused the release of about 650 000 PBq of ^{131}I (2). The major part of this activity was produced in high-yield tests, and was carried upwards into the stratosphere, from which it descended to ground level in a matter of months or years. Consequently, little ^{131}I was available to deliver doses to man. Collective thyroid doses from global fallout of ^{131}I are estimated to have been about 3.3 x 10^6 man Gy, corresponding to a per caput thyroid dose to the world population of about 1 mGy. These figures, however, do not take into account the local and regional collective thyroid doses from the early, low-yield, tests that were conducted in the 1950s. Some of these low-yield tests were carried out at the Nevada Test Site in the United States, mainly from 1951 to 1957; they caused a release of about 5 500 PBq of ^{131}I in the atmosphere. Because the explosive energy of those tests was relatively small, most of the ^{131}I produced remained in the troposphere and was rapidly available to contaminate human foodstuffs. A preliminary estimate of the collective thyroid dose to the population of the contiguous United States from the tests conducted at the Nevada Test Site is about 4 x 10^6 man Gy, corresponding to an average thyroid dose of about 20 mGy for that population (3). The thyroid doses received by the populations living in the vicinity of the Nevada Test Site have been studied in detail (4-6). Individual thyroid doses to children were estimated to range up to 4 600 mGy (5). It should also be noted that the test Bravo, which consisted in the detonation in March 1954 of a high-yield device mounted on a barge over the reef of Bikini Atoll in the mid-Pacific Ocean, resulted in intense local and regional fallout and in the evacuation of about 300 people within 3 days after the detonation as well as the exposure of Japanese fishermen on board the Lucky Dragon (7-8). The estimated thyroid doses which, in this case, were mostly due to short-lived radioiodines, ranged up to 20 000 mGy (9).

- The operation of nuclear reactors leads to relatively small releases of ^{131}I under normal conditions; it is estimated that the total release of ^{131}I from reactors used worldwide for the production of electrical energy up to 1989 was 0.045 PBq and that the corresponding collective thyroid dose to the local and regional populations residing around the reactors is 460 man Gy (2). However, much greater atmospheric releases and doses resulted from the Windscale accident of 1957 and from the Chernobyl accident of 1986. About 0.7 PBq of ^{131}I was released during the Windscale accident (10); the collective thyroid dose was approximately 20 000 man Gy (11), with individual thyroid doses ranging up to 100 mGy (12). During the Chernobyl accident, about 330 PBq of ^{131}I were released into the atmosphere (13); a preliminary estimate of the total collective thyroid dose received by the populations residing in the contaminated areas of the former Soviet Union is about 1.2 x 10^6 man Gy (14); individual thyroid doses up to 10 000 mGy and more are estimated for children in the most affected areas (15). In comparison to those reactor accidents, the reactor accident of Three Mile Island in 1980 was minor with an estimated atmospheric release of ^{131}I of 0.00055 PBq (16-17).

- The operation of plants that reprocess nuclear fuel for civilian purposes results in small releases of ^{131}I because the nuclear fuel originating from the reactor is stored before reprocessing for a time long enough to ensure that ^{131}I has decayed to low levels. It is estimated that the total release of ^{131}I from worldwide civilian fuel reprocessing plants was 0.004 PBq up to 1989 and that the corresponding collective thyroid dose to the local and regional populations residing around the fuel reprocessing plants is 30 man Gy (2). However, much greater atmospheric releases and doses resulted from the operation of plants reprocessing nuclear fuel for military purposes during the early days of the nuclear age when only a short storage time was allowed before reprocessing. Releases of ^{131}I at Hanford from 1947 to 1949 amounted to approximately 25 PBq (18) and a similar figure has been advanced for the releases at Chelyabinsk (19). Assuming a collective thyroid dose of 10^{-11} man Gy per Bq of ^{131}I activity released (2), the collective thyroid doses resulting from the releases from each plant would be about 2 x 10^5 man Gy.

The evaluation of the thyroid doses from radioiodine releases depends on the type of monitoring data that is available. The preferred types of data are, in order of decreasing usefulness: (a) direct thyroid measurements, (b) concentrations in air, milk, and leafy vegetables, (c) depositions per unit area of ground, and (d) atmospheric releases. The use of each type of data will be illustrated in turn.

EVALUATION OF THYROID DOSES BASED ON DIRECT THYROID MEASUREMENTS.

In the case of the nuclear reactor accident that occurred at Chernobyl in April 1986, the content of ^{131}I in the thyroid of people living in the highly contaminated areas of the former Soviet Union was large enough that the gamma radiation emitted by the thyroid as a result of the radioactive decay of ^{131}I could be readily measured with a radiation detector held in the vicinity of the neck. Several hundred thousands people were monitored in this way in Ukraine, Belarus, and Russia within a few weeks after the accident (15; 20-21). The measured exposure rates were then converted into thyroid doses using appropriate assumptions on the dynamics of intake of ^{131}I both before and after the direct thyroid measurement. Since few measurements of ^{131}I in air and foodstuffs were made after the accident in former Republics of the Soviet Union, the dynamics of intake of ^{131}I was largely based on environmental transfer models. Because the ingestion of ^{131}I-contaminated milk was for the majority of individuals exposed the main contributor to the ^{131}I intake (22), the intake function was in most cases represented by the time dependence of the milk contamination following a single deposition.

In the three Republics of the former Soviet Union with highly contaminated areas (Ukraine, Belarus, and Russia), the databases on thyroid doses derived from direct thyroid measurements were used to infer the thyroid doses in areas with few, or no, direct thyroid measurements. In each of the three countries, relationships were sought between the thyroid dose and the ^{137}Cs deposition, which has been extensively measured throughout the contaminated areas:

- in the Chernigov region of Ukraine, the following empirical relationships were obtained (15):

$$D(T) = K \times a^{\exp(-b \times T)} \qquad [1]$$

where: D(T), in cGy, is the thyroid dose for age T (y),
a (unitless) is a parameter representing the thyroid dose at age 0
b (y^{-1}) is a parameter describing the age dependence of the thyroid dose
K (cGy) is a scaling parameter characterizing the radioiodine intake, calculated according to:

$$\log(K) = 640 \times (\log(\sigma(^{137}Cs)) / \rho^2) + 2.7 \times \cos(\phi) + 0.013 \times \rho \times \sin(\phi) - 1.6 \qquad [2]$$

where: $\sigma(^{137}Cs)$ is the ^{137}Cs deposition at the location considered,
ρ (km) and ϕ are the polar coordinates of that location with respect to the Chernobyl site.

- in Belarus, a relationship has been established between the average thyroid dose received by people in rural settlements and the ground-deposition density of radionuclides (^{137}Cs or ^{131}I) in this settlement and in the area around the settlement (20). For the settlements in Hoiniki in Gomel oblast, and Kostukovichi and Krasnopolye in Mogilev oblast:

D_{jx} (adult) = $3.5 \times 10^{-8} \times \sigma_x(^{131}I) + 1.4 \times 10^{-8} \times \sigma_{jx}(^{131}I)$ [3]

$= 3.5 \times 10^{-8} \times R_x \times \sigma_x(^{137}Cs) + 1.4 \times 10^{-8} \times R_{jx} \times \sigma_{jx}(^{137}Cs)$ [4]

where: D_{jx} (adult) is the arithmetic mean thyroid dose, in Gy, for adult population in settlement, j, in area, x, in the absence of any countermeasures in the settlement and for typical lifestyle and dietary habits,

$\sigma_x(^{131}I)$, $\sigma_x(^{137}Cs)$ is the average ground-deposition density, in Bq m^{-2}, of ^{131}I (^{137}Cs), in area, x,

$\sigma_{jx}(^{131}I)$, $\sigma_{jx}(^{137}Cs)$ is the average ground-deposition density, in Bq m^{-2}, of ^{131}I (^{137}Cs), in the settlement, j, in area, x,

R_x, R_{jx} is the average ratio of the ^{131}I to ^{137}Cs ground-deposition densities in area, x, or in the settlement, j, in area, x.

It can be expected that, on average in an area, $\sigma_{jx}(^{131}I) = \sigma_x(^{131}I)$, so that the average relationship between ^{131}I deposition density and the average adult thyroid dose is:

D_x(adult) = $4.9 \times 10^{-8} \times \sigma_x(^{131}I)$ [5]

Ongoing research may clarify whether equations 3 and 4 are generally applicable to all contaminated areas of Belarus. It is likely that correction coefficients taking into account the role of fuel particles, and parameters such as the standing crop biomass, the fraction of cow's intake from pasture grass, and the date of the beginning of cow's pasture will need to be factored in (20).

- in Russia, Zvonova and Balonov (21) analyzed the thyroid measurements and radiation survey data for the Bryansk and Tula oblasts. The thyroid doses derived from direct thyroid measurements for 3 to 6 years old children were found to be correlated with the ^{137}Cs ground-deposition density, the kerma rate in air on May 10-12, 1986 (i.e, 14 to 16 days after the beginning of the accident), the mean ^{131}I concentration in milk in the period May 5-12, 1986, and the average radiocesium content in adults in July-August, 1986. With respect to the ^{137}Cs ground-deposition density, the following relationship was obtained:

$D(3\text{-}6) = (76 \times 10^{-8}) \times \sigma(^{137}Cs)$ [6]

where: D(3-6) is the average thyroid dose for 3 to 6 years old children (Gy), and
$\sigma(^{137}Cs)$ is the ^{137}Cs ground-deposition density (Bq m^{-2}).

According to Zvonova and Balonov (21), the average ratio of the deposition densities of ^{131}I and ^{137}Cs was approximately 8 immediately after the period of intense deposition, so that:

$D(3\text{-}6) = (9.5 \times 10^{-8}) \times \sigma(^{131}I)$ [7]

It should be noted that the measurement of the gamma radiation emitted by the thyroid with a radiation detector held in the vicinity of the neck is not the only method available to derive the thyroid dose from a direct measurement on man. Measurements of ^{131}I in urine also have been used for that purpose, notably to estimate the thyroid doses received by the Marshallese as a result of the test Bravo, in 1954 (9).

EVALUATION OF THYROID DOSES BASED ON MEASUREMENTS IN FOODSTUFFS AND IN AIR.

For regions other than the most contaminated areas of the former Soviet Union, the evaluation of the thyroid doses resulting from the Chernobyl accident was based on measurements in foodstuffs and in air. In Russia, Kryshev (22) made use of the detailed data of radioecological monitoring that were available for Sosnovy Bor near Leningrad to infer that the thyroid doses from inhalation were much lower than those due to ingestion and that the main contributors to the thyroid dose received by children were the consumption of milk, followed by the consumption of water, vegetables, bread, and home-baked goods. The following empirical relationship was derived (22):

$D(T) = K(T) \times R(T) \times \sigma(^{131}I)$ [8]

where: D(T) is the thyroid dose, in Gy, for age, T, in years,
R(T) is the thyroid dose coefficient, in Gy Bq^{-1},
$\sigma(^{131}I)$ is the ^{131}I deposition density, in Bq m^{-2}, derived from the ^{137}Cs ground-deposition density, and calculated for 15 May 1986 (about 17 days after the likely time of highest deposition),
K(T) is the ^{131}I transfer coefficient from ground deposition to intake by people of age, T, in Bq per Bq m^{-2}.

The following values of K(T) were obtained by Kryshev (22):

T (y)	1	5	10	15	adult
K (Bq per Bq m^{-2})	0.28	0.32	0.36	0.36	0.47

When recalculated to reflect the transfer coefficient related to the total ground deposition of ^{131}I, assumed to have occurred on 28 April 1986, the values of K(T) become:

T (y)	1	5	10	15	adult
K (Bq per Bq m^{-2})	0.065	0.074	0.083	0.083	0.11

On the basis of the variation of K with age shown above, using the thyroid dose coefficients recommended by ICRP (23), and population fractions of 0.02, 0.16, 0.20, and 0.62 for the age groups around 1 y, 5 y, 10 y, 15 y, and for adults, respectively, a transfer coefficient from ^{131}I deposition density to per caput thyroid dose of 7 x 10^{-8} Gy per Bq m^{-2} is obtained.

The same parameter values can also be used to derive the per caput transfer coefficients from the adult values for Belarus (equation 5) and from the 3 to 6 years old children for Russia (equation 7; data from Zvonova and Balonov). The per caput transfer coefficients obtained in that way are 7 x 10^{-8} Gy per Bq m^{-2} for Belarus and 5 x 10^{-8} Gy per Bq m^{-2} for Russia (data from Zvonova and Balonov).

The ^{131}I concentrations in milk and in leafy vegetables that were measured in most countries of the Northern hemisphere after the Chernobyl accident were used by the UNSCEAR Committee to estimate in a consistent manner the thyroid doses received by the populations of those countries (13). The ^{131}I concentrations in leafy vegetables were found to be, on average, 1.5 times greater than those in milk. However, when the relative consumption rates of milk and leafy vegetables are taken into account, the ^{131}I intakes from milk were, on average, much greater than those from leafy vegetables, especially in the countries of Northern, Central, and Eastern Europe that were most affected by the Chernobyl accident.

Measurements of ^{131}I concentration in milk were also used by the UNSCEAR Committee to estimate the thyroid doses due to global fallout from the nuclear weapons tests conducted in the atmosphere, mainly in the 1960s (2; 17). On the basis of measurements in Argentina (24), the relationship between ^{131}I ground-deposition density, $\sigma(^{131}I)$ in Bq m^{-2}, and time-integrated milk concentration, C_m in Bq a L^{-1}, was estimated to be:

$$C_m = 6.3 \times 10^{-4}\ \sigma(^{131}I) \qquad [9]$$

A transfer coefficient from ^{131}I deposition density to per caput thyroid dose of 8 x 10^{-8} Gy per Bq m^{-2} is derived from equation 9 using the values adopted by UNSCEAR (2) of 0.3 L d^{-1} for the average milk consumption rate and 1.2 x 10^{-6} Gy Bq^{-1} for the age and milk consumption weighted thyroid dose coefficient.

EVALUATION OF THYROID DOSES BASED ON MEASUREMENTS OF GROUND DEPOSITION.

The importance of the deposition - pasture grass - cow's milk pathway to man as a major dose contributor in the case of an atmospheric release of ^{131}I was not fully recognized until 1957. Therefore, the dose reconstruction efforts related to radioiodine releases prior to 1957 are largely based on measurements of radionuclides other than ^{131}I (or of total beta measurements or exposure rates) as well as on environmental transfer models.

Atmospheric testing of nuclear-weapons-related devices at the Nevada Test Site (NTS) began in 1951; most of the atmospheric releases of radioactive materials, including ^{131}I, took place in test series conducted in 1951, 1953, 1955, and 1957. In the Off-Site Radiation Exposure Review Project of the U.S. Department of Energy, doses were estimated for the "local" populations (less than 800 km from the NTS); the key data used to reconstruct thyroid doses from ^{131}I releases were estimates of ground-deposition densities of ^{131}I from each location of interest and each important test, which were derived either from the available exposure rates or from contemporary measurements of ^{137}Cs and $^{239,240}Pu$ in soil, combined with data on the relative abundance of radionuclides produced in each test (25-27). The transfer coefficients representing the ^{131}I intakes per unit deposition density were estimated to vary according to the fallout date and to the age and sex of the person considered (28). Examples of results, averaged over both sexes, for a date with low transfer coefficient (17 March) and for a date with high transfer coefficient (24 July) are (28):

Age (y)	<1	1-11	12-18	Adult
Transfer coefficient (Bq per Bq m^{-2}): fallout on 17 March	0.0044	0.0092	0.013	0.016
Transfer coefficient (Bq per Bq m^{-2}): fallout on 24 July	0.17	0.14	0.13	0.08

The per caput thyroid doses per unit deposition density, calculated using the thyroid dose coefficients recommended by ICRP (23), are found to vary from 9×10^{-9} to 9×10^{-8} Gy per Bq m^{-2} according to the date of fallout.

The U.S. National Cancer Institute also is carrying out a study related to ^{131}I releases from the NTS; that study includes the estimation of thyroid doses received by populations across the continental United States (29). According to preliminary estimates, the amount of ^{131}I that was deposited throughout the continental United States as a result of the weapons tests conducted at the NTS is 1.5 EBq (over an area of 7.7×10^6 km^2) and the per caput thyroid dose was about 20 mGy, corresponding to an average thyroid dose per unit deposition density of about 10^{-7} Gy per Bq m^{-2} (3).

EVALUATION OF THYROID DOSES BASED ON MEASUREMENTS OF AIRBORNE RELEASES.

There are cases where human or environmental radiation monitoring data are not available, either because the radioiodine releases, although important, occurred during the early days of the nuclear age, when environmental monitoring was not a priority, or because the radioiodine releases are so low that they cannot be readily detected in the environment. In the absence of human or environmental monitoring data, the evaluation of the thyroid doses is based on estimates of radioiodine releases.

For example, the ^{131}I releases from the Hanford Site in the United States for the years 1945 to 1951 were estimated on the basis of the operating histories of facilities on the site and on the knowledge of the effluent control technologies in use at the time of release (18). Detailed thyroid dose estimates were then prepared for 1102 locations around the site, 12 different representative individuals, distinguished by age and gender, and a series of food source scenarios, by means of environmental transfer models and data on milk production, distribution and consumption, among other factors (30).

Routine reactor releases have been usually so low in recent years that the environmental concentrations are not detectable and the resulting thyroid doses can only be calculated by means of environmental transfer models. The UNSCEAR Committee, which compiles every few years the data available on routine releases from nuclear power plants and other nuclear facilities (2; 13; 17), uses a model site and model calculations to evaluate the collective doses resulting from routine releases of radionuclides; the collective thyroid dose per unit of ^{131}I activity released is estimated by the UNSCEAR Committee to be about 10^{-11} man Gy per Bq (2; 13; 17).

DISCUSSION.

General information on the major sources of radioiodine releases into the atmosphere from the nuclear industry is summarized on Table 1. Most of those releases occurred more than 30 years ago. The nuclear weapons tests that were conducted in the atmosphere account for most of the ^{131}I that was released into the environment and for most of the resulting collective thyroid dose.

The thyroid doses were evaluated using different methods according to the types of radiation data that were available. Table 2 presents values of collective thyroid doses normalized per unit of activity released and of average thyroid dose normalized per unit deposition density. As expected, the normalized collective doses are much greater for ground-level releases than for the nuclear weapons tests, which carried the radioactive materials that they produced to high altitudes. The average thyroid doses per unit deposition density are remarkably similar, given the variety of methods and models that were used to estimate that quantity. It should be kept in mind, however, that for a given deposition density, the individual thyroid doses vary by orders of magnitude according to the date of fallout, agricultural practices, the age of the person considered and dietary habits.

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Table 1. Major sources of radioiodine releases into the environment.

	Activity released (PBq)	Year(s) of release	Collective thyroid dose (man Gy)	Main basis for thyroid dose evaluation
MILITARY NUCLEAR FUEL CYCLE				
Atmospheric nuclear weapons tests:				
• Global fallout	650 000	1962-1963 (mainly)	3.3×10^6	^{131}I deposition densities and concentrations in milk
• Local and regional fallout (Nevada Test Site)	5 500	1951-1957 (mainly)	4×10^6	^{131}I deposition densities
Reactors:				
• Windscale accident	0.7	1957	2×10^4	^{131}I release and concentrations in milk
Fuel reprocessing plants (routine releases):				
• Hanford	25	1945-1948 (mainly)	2×10^5	^{131}I release
• Chelyabinsk	about 20	1949-1951 (mainly)	2×10^5	^{131}I release
CIVILIAN NUCLEAR FUEL CYCLE				
Reactors:				
• Routine releases	0.045	up to 1989	460	^{131}I release
• TMI accident	0.00055	1980	small	^{131}I release
• Chernobyl accident	330	1986	1.2×10^6	Direct thyroid measurements and ^{137}Cs deposition densities

Table 2 - Transfer coefficients

COLLECTIVE THYROID DOSE PER UNIT ACTIVITY RELEASED (man Gy per Bq)	
Global fallout from weapons tests	5×10^{-15} (world population)
Local and regional fallout from weapons tests (Nevada Test Site)	7×10^{-13} (U.S. population)
Reactor accident at Windscale	3×10^{-11}
Reactor accident at Chernobyl	4×10^{-12}
Routine releases at ground level	1×10^{-11}
AVERAGE THYROID DOSE PER UNIT DEPOSITION DENSITY (Gy per Bq m^{-2})	
Global fallout from weapons tests	8×10^{-8}
Local and regional fallout from weapons tests (Nevada Test Site) (3)	1×10^{-7} (U.S. population)
Local fallout from weapons tests (Nevada Test Site) (28)	from 9×10^{-9} to 9×10^{-8} (local population)
Reactor accident at Chernobyl: Belarus (20)	7×10^{-8}
Reactor accident at Chernobyl: Russia (21)	5×10^{-8}
Reactor accident at Chernobyl: Russia (22)	7×10^{-8}