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Oral intake of radionuclides in the population

A review of biological factors of relevance for assessment of absorbed dose at long term waste storage

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Umeå, Sweden, October 1982

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

A list of other reports published in this series during 1982, is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26) and 1981 (TR 81-17) is available through SKBF/KBS.

REVIEW OF DOSE FACTORS FOR ORAL INTAKE OF RADIONUCLIDES OF POTENTIAL HAZARD TO THE POPULATION FROM LONG TERM STORAGE OF HIGH LEVEL WASTE.

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Abstract

Dose factors of some radionuclides have been reviewed with respect to a chronic oral intake by members of the public. The radionuclides taken into account are Pu-239, Np-237, Ra-226, Th-230, Pa-231, Tc-99 and I-129, all of which might be of potential hazard at a long term storage disposal.

The parameter that have the major influence on the dose factor, for most of the radionuclides studied, is the uptake from the gut. In order to assess the dose factor it is therefore essential to make a good estimate of the gastrointestinal uptake of the radionuclides under the actual conditions. The "annual limit of intake" (ALI) given in ICRP 30, is intended to be applicable on a population of workers, and for a single intake. Since the gut uptake figures in the ICRP-publication are based mainly on uptake values recieved in experimental animals, given single relatively large oral doses of the isotope studied.

From a review of current litteratur, gut absorbation factors and dose factors, to be used for members of the public at a chronic oral intake, are suggested. Compared with those for workers in ICRP 30, the dose factors increases for plutonium and protactinium, and decreases for neptunium. An attempt to predict possible future changes of the ALI for members of the general public is also made.

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1 Introduction

Evaluations of potential dangers from buried high level radioactive waste nearly always include contamination of food or water supplies through seepage from the storage site. These factors are of dominant importance. Safety analyses therefore depend to a large extent on the ingestion hazard associated with the waste.

The radionuclides of interest in this report, are those with a long physical half-life, which will make a major contribution to the radiation risk in the distant future, or radionuclides for which the present risk estimate is uncertain due to a lack of data. The radionuclides chosen are the actinides plutonium-239, neptunium-237, protactinium-231 and thorium-230, as well as radium-226 and the fission products technetium-99 and iodine-129. The aim of this report is to present a more reliable basis for calculation of the Committed Dose Equivalent to a member of the population due to oral intake in a form that might be expected after leakage from the waste disposal facility.

The radionuclides included usually have radioactive daughters; these are also included in the dose calculations. The decay chains are illustrated in appendix 1.

2 ICRP activities concerning radionuclide intake and metabolism.

2.1 The concept of effective dose equivalent

The International Commission on Radiological Protection (ICRP) recently introduced the concept of "effective dose-equivalent" (ICRP77, ICRP78). This is a weighted average of committed dose equivalents for specific organs. The weights are associated with the respective stochastic risk factor. The risk of fatal cancer or serious hereditary disease in children and grandchildren is taken into consideration. It should thus be possible to compare the risk from irradiation with other risks. The risk factors were estimated by the ICRP based on material compiled by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), which published its most recent report in 1977 (UN77).

With this concept in mind, the earlier dose-equivalent limits for a number of organs and tissues can be replaced by a single figure. This figure should be augmented by a limit based on non-stochastic effects, such as opacity of the lens.

ICRP 26 recommends the quality factor Q=20 to be used for alpha particles, and Q=1 for all photons and electrons. The quality factor is intended to allow for the biological effectiveness of the microscopic distribution of the absorbed energy. The absorbed dose should be multiplied by Q to obtain the absorbed dose equivalent.

Table 2.1. Weight factors for calculation of the effective dose-equivalent.

Tissue	w_T
Gonads	0.25
Breast	0.15
Red bone marrow	0.12
Lung	0.12
Thyroid	0.03
Bone surface	0.03
Remainder	0.30

The remainder is divided equally between the five remaining organs or tissues receiving the highest dose equivalent.

2.2 Annual limits on intake

The publication of new basic recommendations in ICRP publication 26, together with a better understanding of uptake and retention of radionuclides in body tissues, and of the decay scheme of the radionuclides, requires new exposure limits for radionuclides. Recommended values for maximum permissible concentrations in air and water and maximum permissible body burdens were published in ICRP publication 2 (ICRP59), adopted in 1959. This publication was recently superseded by publication 30, published in three volumes (ICRP79, ICRP80, ICRP81). This publication presents limits on the exposure of workers in the form of Annual Limits on Intake (ALI) for the oral and inhalation routes of entry into the body. The ALI is calculated on the basis of the recommendation in ICRP 26. With respect to stochastic effects (cancer induction and genetic damage), the effective dose equivalent must not exceed 50 mSv, and the absorbed dose equivalent to a single organ must not exceed 500 mSv, nonstochastic effect. The annual intake may therefore be limited by different effects for different radionuclides .

2.3 ICRP metabolic models

2.3.1 ICRP 30

For the purpose of calculating the annual limits on intake, ICRP 30 also presents metabolic data for all elements. In part one, dosimetric methods are described, and dosimetric models for the respiratory system and the gastro-intestinal tract are presented, as well as a dosimetric model for bone. Details of the gastrointestinal model and the bone model will be discussed later. References are given for the metabolic models. In some cases, the models can be quite detailed and can be supported by a great deal of experimental evidence from observations on humans. In other cases, data can be limited to a few observations on a single species of experimental animal. This lack of data on metabolism represents the largest factor of uncertainty in most estimates of the absorbed dose.

It should be noted that ICRP 30 deals with occupational exposure of individuals, not with exposure of ordinary members of a population. It is therefore possible

that the metabolic model is more suitable for the former than the latter group. It describes the metabolism of the radionuclide after a single relatively large intake of an inorganic form. The dose equivalent is integrated over a period of 50 years.

2.3.2 ICRP 19, 20 and 11

In 1972, the ICRP published two reports, one providing a comprehensive review of the metabolism of compounds of plutonium and other actinides (ICRP72), the other of alkaline earths (ICRP73). The former report deals mainly with plutonium, in the inhalation case solely with plutonium. However, some data are discussed for other actinides as well, especially americium and curium, but thorium, actinium and neptunium are also mentioned in the report. Protactinium and uranium are not mentioned. Radium, being an alkaline earth, is included in the latter report.

The ICRP has also published a report entitled "A review of the radiosensitivity of the tissues in bone" (ICRP68), where the deposition pattern in the skeleton for some radionuclides is studied in detail.

2.4 Models for the gastro-intestinal tract

The dosimetric model used by the ICRP in their publication 30 (ICRP79) is essentially the one conceived by Eve and Dolphin (Ev66, Do66) as modified by Bernard and Hayes (Be70) in their "catenary compartment model". It comprises 4 sections: stomach, small intestine, upper large intestine and lower large intestine The mean residence time adopted is 1 hour in the stomach, 4 hours in the small intestine, 13 hours in the upper large intestine and 24 hours in the lower large intestine. Excretion from the different parts is assumed to be exponential, and within each compartment immediate mixture is assumed. Absorption from the gut is assumed to take place solely in the small intestine. The model is intended to be applicable to a population of radiation workers.

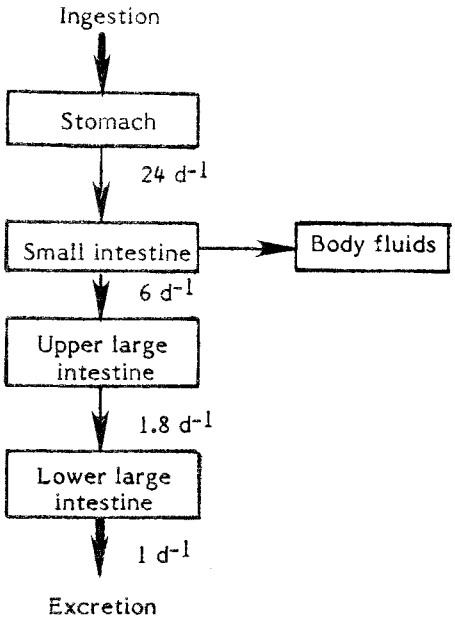

Figure 2.1. ICRP 30 metabolic model of the gastro-intestinal tract.

This is obviously an approximation:

1. The contents do not pass the intestine at a uniform rate. The flow rate is higher at the beginning and lower at the end of each compartment, due to absorption of material. The flow rate also varies with time.

2. The material is not completely or immediately mixed in the different compartments; the movement more resembles flow through a tube.

3. The model does not take into account the fact that absorption can take place over different parts of the small intestine.

A more refined dosimetric model has been introduced by Skrable et al. (Sk75). They use the same physiological model as a basis (Ev66). They assume that ingested material instantaneously mixes with the contents of the stomach and is transferred to the small intestine at an instantaneous fractional rate of 1.0 h^{-1}. For the remainder of the GI-tract they let the contents move through the different segments in a slug flow fashion. This assumption makes calculation of the dose much more complicated for penetrating radiation (X-rays or gamma-

rays with an energy of more than 10 keV). (This problem is currently being studied by the ICRP Task Group on Reference Man). The advantage of this model is that a point for the maximum absorbed dose can be established, and the dose at that point calculated. Compared to the very great individual variations of the physiological parameters, the absorbed dose and, hence, the risk, to the walls of the GI-tract can be determined with sufficient accuracy with the ICRP model.

Vacca and coworkers (Va68) have also developed a mathematical model for the dosimetry of the GI-tract. With their method, the cumulated activity at any point of the GI-tract can be determined, and thus the dose to any small element can be limited.

Another obvious method for refining the ICRP dosimetric model is to divide the GI-tract into more sections than 4. This also provides more detailed information on the distribution of the cumulated activity within the GI-tract. However, as was the case with the "slug flow" model, one gets into trouble in calculating the absorbed dose from penetrating radiation.

The depth where the sensitive layer of the gastro-intestinal mucosa is found varies from 0.14 mm in the small intestine to 0.28 mm in the stomach and 0.42 mm in the rectum (Ev66). Layers of non-mitosing cells are found at more shallow depth. The epithelium on the internal surface of the GI-tract is constantly being worn away and renewed after a few days from below. The absorbed fraction to the mucosal sensitive layer is a factor of between 0 and 1. For non-penentrating radiation, the specific absorbed fraction in the mucosa is $1/2 \times v/M_C$, where M_C is the mass of the contents of the section regarded and v is a factor of between 0 and 1 representing the degree to which the radiation penetrates the mucus and reaches the sensitive layer. The factor 1/2 is introduced because the dose at the surface of the contents is about one half of the dose within the contents, since the range of the beta or alpha particles is much less than the diameter of the filled intestine. For beta-radiation, v is taken to be 1, and for alpha, the ICRP sets v equal to 0.01.

Thorne recently presented a modification of the ICRP GI-model (Th80b). The reason for this is that experimental data have been obtained that conflict with the ICRP model. Sullivan (Su80b) reported data on the distribution of actinides between the contents of the gastrointestinal tract and the wall for neonatal rats. In these animals, the quantity of activity in the walls of the tracts, 7 days after administration is found to be typically at least as large, and sometimes an order of magnitude larger, than the quantity of the actinide in all other organs and tissues of the body. Together with other reported prolonged retention of cerium and ruthenium in the intestine (Mat69, In72, Su78), this indicates that there are two processes involved in the absorption of these radionuclides from gut to blood: 1. internalization within the mucosa; 2. transport from the intestinal mucosa into the blood. Some of the internalized radioactivity can eventually be sloughed into the lumen and excreted. On this basis, Thorne (Th80) has presented a revised gastrointestinal tract model, which incorporates the wall as a compartment. For the contents of this compartment, v in the equation above increases for alpha particles from 0.01 to 1.0. This increase is of little significance for the effective dose equivalent when the fraction transferred to the blood is very small. In young animals, however, fractional transfers of actindes in excess of 0.01 have been reported in some cases (Su80a), and in these cases the use of the ICRP model might give rise to a significant underestimation of the effective dose equivalent. The effect will be more pronounced for more short-lived radionuclides.

2.5 ICRP dosimetric model for bone

Bone is a highly complex and heterogeneous tissue with cellular and mineral components arranged in a varied and irregular pattern. This means that one encounters special problems in the dosimetry of radionuclides deposited in the skeleton. The absorbed dose equivalent, especially from alpha and beta radiation, will vary widely within the skeleton depending on the site of deposition and the microscopic structure of the bone tissue at that point.

In the ICRP dosimetric model for the skeleton the bone surface has been recognized as the most radiation-sensitive part of the mineral bone (ICRP68, ICRP78). The surface is assumed to have a thickness of 10 μm and a mass of

120 g. The mass distribution between trabecular and cortical bone is 0.5 : 0.5 when only the surface is considered. If the total volume is taken into account, the distribution is 0.2 : 0.8. When the absorbed dose to the skeleton or the bone marrow is calculated, the distribution of the radionuclides within the skeleton should be taken in consideration. For most elements, knowledge of this distribution is sorely lacking. The ICRP has therefore issued the general recommendation that radionuclides with a physical half-life of less than 15 days should be considered as surface deposited; for the rest, nothing is said (ICRP78).

A surface distributed radionuclide will deliver a greater absorbed dose to the red marrow than a volume distributed one. This is especially the case for radionuclides emitting alpha or beta radiation. For this type of radiation, with its source in trabecular bone, the ICRP recommends an absorbed fraction to the red marrow of 0.5 when the source is surface distributed, and 0.05 and 0.35, respectively, with a volume-distributed source. No beta or alpha particles can reach the red marrow from cortical bone. For alpha-emitting radionuclides and beta-emitting radionuclides with low beta-energy ($E < 0.2$ MeV), the absorbed dose to the bone surfaces will be 10 times higher for a surface deposition than for a volume deposition, according to the absorbed fractions recommended in ICRP 30. For beta-emitters with higher energy, there is no significant difference. For photons, the absorbed dose to the bone surfaces is roughly equal to the absorbed dose in the total skeleton, with good approximation.

2.6 The applicability of ICRP 30 - models and limits of intake

As is stated in ICRP 30 (ICRP79), the published ALI values are limits derived from given exposure limits for occupationally exposed adults (ICRP77) and effective dose equivalents calculated from the metabolic data adopted. The commission does not recommend the data and the models described in ICRP 30 for estimation of the committed dose equivalent to members of a population by adjustment solely on the basis of difference in mass of organs. The basis for this recommendation are the following reasons.

- A population has a different distribution according to age and sex than occupationally exposed workers. This means a different distribution of radiation sensitivity.

- The metabolic data chosen are usually based on studies where a relatively large intake of a radioactive substance was administred in a single dose. Small and chronic intake might sometimes result in a different distribution and different uptake factors.

- Binding of the substance to food and water in low concentrations will affect the absorption factor in many cases. Reduced hydrolysis in the small intestine due to formation of stable complexes in food material may lead to increased absorption, although it is also possible that uptake could be limited as a result of strong binding to poorly absorbed substances.

- Children have a different metabolism than adults; retention time in the body is usually shorter. In infants, gastrointestinal uptake is also often several orders of magnitude higher.

- The weight factors used in calculating the effective dose equivalent are based on data for adults. However, the ICRP has stated that, in lieu of better data, they may also be used for children (ICRP77). There are some obvious differences between children and adults on this point, e.g. the genetic effect, which should be of greater importance in children.

2.7 General uncertainty in the ICRP 30 models, and ALI

2.7.1 Models

Even an ideal metabolic model that accurately describes the metabolism of the radionuclide in the average man will be subject to wide individual variation. The factor for which this individual variation might cause the greatest variation in the dose conversion factor is gut uptake. Laboratory animals are used to assess average gut uptake, since human data are not available at present for most of the radionuclides. The laboratory animals used are often small ones, rats or

hamsters. These are rodents, and as such they have a different gastrointestinal environment. Rather large differences are often found between the species e.g. hamsters are known to absorb less plutonium than rats by a factor of two to three (Ha82b). Also, the animal's feeding pattern has been found to influence radionuclide absorption; animals who have been starved for a while usually absorb poorly absorable radionuclides to a higher degree (Ha82b). This means that more is absorbed from an empty stomach and small intestine. Man ususally empties his stomach before eating the next meal, while the laboratory animals normally seem to eat continously. The higher degree of absorption in starved animals indicates that the eating pattern of man might increase absorption. A general approximation that has been made in the models is that daughters remain at their place of formation and follow the metabolism of the original radionuclide. In many cases, this approximation is a minor one, but in some cases, when the physical half-lives are favourable, redistribution might significantly change the absorbed dose.

2.7.2 Annual Limit of Intake

The Annual Limit of Intake (ALI) for occupationally exposed persons depends on 1) the chosen system of dose limitation, 2) the physical properties of the radionuclide and its daughters, 3) the relative biological effectivness (RBE) of the radiation emitted by the radionuclide, and 4) the metabolic behavior of the radionuclide compound. It will also depend on approximations made for calculation purposes, since it is based on the concept of "effective dose equivalent", the value of the ALI will be fairly insensitive to changes in biological distribution.

These are several reasons for a possible future change of the ALI for a certain radionuclide compound.

1. The f-value (gastro-intestinal uptake) is changed.
2. A new distribution within the body is adopted.
3. A new biological half-life, or excretion pattern, is adopted.
4. The quality factor, Q, is changed due to changed RBE.
5. The weight factors, which is based on risk estimation, are changed.

6. The given limits of the committed dose equivalent are changed.
7. New physical properties of the radionuclide are discovered.
8. The models for absorbed dose calculation are improved.

Some of these factors will probably only have a minor influence on the ALI, especially those associated with physical parameters. The major factor is generally 1, but may also be some of the others.

3 Variation of absorbed dose and committed dose equivalent with age

Besides the physical properties of the radionuclide and the distribution and metabolism of the radiactive substance within the body, the absorbed dose is dependent on the anatomic make-up of the organism, i.e. organ mass and distances between organs.

Penetrating radiation (photons with energy > 10 keV) stemming from a source organ will irradiate other organs as well. In children, whose body size is smaller, the radiation will reach more distant organs than in adults, which results in a higher absorbed dose in those organs. Similar, absorbed doses from non-penetrating radiation (low-energy photons, electrons, beta- and alpha-particles), which is absorbed solely in the same organ from which they originated, are inversely proportional to organ mass and thus show considerable variation with age.

Generally the child's intake of food and water is smaller than that of the adult, and similarly, the child's higher rate of metabolism often leads to a more rapid elimination of activity from his body than would be typical for adults. The higher absorbed dose that is received by a child due to the smaller size of its body is thus to some extent compensated for by this effect.

The expected longer remaining lifetime for a child than for an adult must be taken into account when calculating the committed dose equivalent resulting from an intake in childhood. For radionuclides with long biological and physical retention times, it is generally not enough to integrate for 50 years, an integration for 70 years should instead be performed. In these cases, allowance should also be made for the fact that the time the subject remains a child might be small compared to the total time of exposure.

Of the radionuclides included in this report, a significant difference in the committed dose equivalent due to different body size can only be expected for technetium 99 and iodine-129. This difference can roughly be estimated to be about a factor of 10 higher dose equivalent for a newborn than for an adult, a factor of 5 for a five-year-old child and a factor of 2 for a 10-year-old.

For radium and the actinides included in this report, smaller body size during childhood has only a minor influence on the committed dose equivalent. The important factors for these radionuclides are: 1) the additional absorbed dose accumulated in the 50-70 year period, and 2) the usually higher gut uptake of these substances in the newborn child. The first factor can easily be calculated on the basis of the metabolic model. This has been done by Ryan and Dunning Jr. (Ry81). For the radionuclides in question, the results are presented in table 3.1 together with the results of my own calculations.

Increased gut uptake is preferentially noted in suckling children; after weaning, uptake is comparable with that in the adult. Increased uptake for infants is generally estimated to about a factor of 100, except for neptunium, where a factor of 10 seems to be a more accurate estimate of the increase of the ICRP 30 value. In evaluating the significance of this higher gut uptake cognizance should be taken of the fact that the milk diet of these infants is considerably less contaminated with these radionuclides (with the possible exception of radium) than ordinary human foods.

Table 3.1. Per cent increase in committed dose equivalent to limiting organ and effective dose equivalent due to integration over 70 instead of 50 years.

	Committed Dose Equivalent to limiting organ		Effective Dose Equivalent	
	This report	Ryan & Dunning	This report	Ryan & Dunning
Pu-239	31	31	25	25
Np-237	32	32	29	29
Ra-226	-	10	-	9
Th-230	14	32	10	29
Pa-231	50	46	51	42

4 Transfer to fetus and infants

4.1 Cross-placental transfer

Already in 1953 the placental transmission of radioactive alkaline earths was studied in pregnant rats by Wilkinson and Hoecker (Wi53). For plutonium, they reported that about 0.002 % to 0.03 % of the intraveneously injected activity was found in the fetus 5 days later. They also observed that with an increase in the amount of plutonium administred to the mother, there is a corresponding decrease in the amount which the placenta will transmit to the fetus. They draw the conclusion that there is no active transfer of plutonium across the placenta barrier, since the placental level of the plutonium was higher than the corresponding fetal value. The same authors also reported studies with radium, but these values are presented in a way that makes them more difficult to interpret.

Subsequently, investigations of placental transfer of plutonium in rats, guinea pigs and baboons have been presented (Si76, Si78, Su80c). The results from these studies are generally similar between the different animal species. For rats 0.01 - 0.04 % of injected plutonium is found in the fetus (Si76), and for baboons Sikov et al. found a relative concentration in the embryo of 0.01 - 0.1 compared to the mother (Si78). Sullivan has studied the accumulation of plutonium **in utero** in relation to the difference in development of immunity; no such relationship was found, however (Su80c). Moreover, specific activity has been found not to be an important factor in the cross-placental transfer of plutonium (Si76).

The placenta barrier is ineffective for radium; the same concentration is built up in the fetus as in the mother (Wr77).

Human data is available for iodine. Uptake in the fetus is dominated by enrichment in the thyroid and starts around the 12th week. Dyer (Dy69) measured fractional uptake in the fetus at different fetal ages, table 4.1.

Table 4.1. Uptake of iodine-131 in fetus (Dy 69).

Fetal age (weeks)	Fraction of administrated activity found in fetus %
11	0.003
14	0.06
15	0.08
15	0.12
22	1.37

4.2 Transfer to suckling children

Except for iodine and technetium, which have radioisotopes that are used medically, no human data exists concerning milk excretion, and animal data is sparse.

One article has been found dealing with plutonium and neptunium (Mc62). The authors of this article are interested in the hazard to man resulting from the ingestion of the milk of animals grazing on contaminated forage. They measured the plasma to milk ratio following an intraveneous injection in a mature sheep. They found an average concentration ratio of milk to plasma of 0.025 for plutonium, and 0.05 for neptunium. An ability to discriminate between these radionuclides is thus demonstrated by the milk excretion mechanism. Since excretion of deposited plutonium and neptunium is very slow, these substanses will be found in the milk only immediately after uptake from the gut. Assuming a plasma volume of 2500 ml (ICRP75), a daily milk consumption by the child of 850 ml (Ma81) and a half-life in plasma of 0.25 d (ICRP79), an average of 0.3 % of the plutonium and 0.7 % neptunium absorbed by the mother can be estimated to be excreted via the breast milk.

Unfortunately, no milk excretion data have been found on radium, but since radium is a calcium analog, a significant fraction can be expected to be excreted via the breast milk. A milk:plasma concentration ratio of 35.0 is reported for calcium-45 (Mc62).

For iodine-131, a study by Larsson et al. showed a total excretion via the breast milk of 5 % of administred activity (La76). This value should also be applicable to iodine-129.

A fraction of 20 % of technetium-99m pertechnetate has been found to be excreted via the breast milk in human studies (Ma82). This value, which is remarkably high, is probably also the best estimate for technetium-99 of environmental origin.

5 Review of the specific radionuclides

5.1 Plutonium-239

Plutonium-239 has a half-life of 24000 years. The element can exist chemically in oxidation states VI, IV and III.

Plutonium is the most extensively studied element, with respect to metabolism, of those included in this report. As a result of nuclear weapons testing, measurable amounts of plutonium can be found in different human tissues. Before 1975, this was due mainly to inhalation (Be76), but since the atmospheric concentration of plutonium is lower now, ingestion may account for a relatively higher proportion of plutonium intake. No direct measurements have been made of gastrointestinal absorption of plutonium in humans. Some human data are, however, available on plutonium metabolism, although most information is based on animal studies.

Surveys concerning the applicability of existing metabolic data to the general public, and of the influence of environmental factors on gut absorption, have recently been published by Harrison et al. (Ha81b, Ha82a). Thompson has also used plutonium as an example in an attempt to derive ALI values for members of the public from the data in ICRP 30 (Th80a).

5.1.1 ICRP 30 metabolic model

The metabolism of plutonium was reviewed in 1970 by an ICRP task group. The results were published in ICRP report 19 (ICRP72). Recently, however, an ICRP task group was established to review this report (No81). The 1972 report recommended a value of 0.003 % as an uptake factor. Several recent investigations did, however, indicate somewhat higher absorption, and when ICRP 30 was published, the f factor for soluble plutonium was increased by a factor of

three to 0.01 % (ICRP79). For insoluble compounds of plutonium, oxides and hydroxides, the uptake is 0.001 %, which is higher by a factor of ten than the value recommended in ICRP 19. This figure is based mainly on a study by Stather and coworkers (St79).

In ICRP 19 plutonium absorbed into the blood stream is assumed to be distributed as follows: 45 % in bone, 45 % in the liver and the remaning 10 % homogeneously distributed in all other tissues and early excreta. A biological half-life of 100 years in the skeleton and 40 years in the liver is suggested. ICRP 30 adopted these values for bone and liver, but the remaining 10 % is assumed to be directly excreted, except for a fraction deposited in the gonads. This fraction corresponds to a concentration of 0.001 % per gram of gonadal tissue, is a six-fold higher activity concentration and is assumed to be permanently retained. The plutonium deposited in the skeleton is assumed for the purpose of dosimetry always to be found on the endosteal surfaces of the bone.

The annual intake of plutonium-239 is limited by the absorbed dose equivalent to bone surfaces.

5.1.2 Reliability of the ICRP 30 model

This metabolic model is conservative on the following points.

- Extrapolating retention half-lives in the liver and skeleton from small animals to man according to a power function of the body weight appears to result in a conservative dose estimate (Ro81). The adopted half-life is a conservative estimate; no experiments have been conducted with animals long-lived enough to confirm the figure of 100 years in the skeleton.

- With the chosen distribution figures, the dose to the liver and skeleton can only be underestimated by a factor of two.

- The assumption that plutonium in the skeleton is fixed on the bone surfaces at all time after deposition is conservative, as plutonium is buried and recycled

in the bone. Burying is a process that substantially reduces the committed dose equivalent to the bone. Only the dose to the surfaces is taken into account in calculating the effective dose equivalent, which is therefore considerably overestimated. Priest and Hunt have developed a model which allows for plutonium burial and recycling (Pr79). Compared to their model the ICRP "surface model" overestimates the absorbed dose equivalent to the bone surfaces, which is the limiting organ, by a factor of four.

- The results of animal studies indicate that there is no selective accumulation of plutonium or other actinides by the gonads. The higher concentration is assumed in lieu of better data. The studies indicate, however, that any activity deposited in them is retained there indefinitely (Ri75,Ne79). Even if the ICRP 30 fractional deposition of plutonium in the gonads is based on the data reviewed by Richmond and Thomas (Ri75), they adopt a value which is about five times higher than that recommended by these authors. Richmond and Thomas also report that measurements of the concentration of fallout plutonium in humans show values in the gonads similar to those in other soft tissue.

Available data from human autopsies suggest that the average fraction of plutonium deposited in the skeleton is 45 % (Mc76). The range of values found in autopsy studies runs from below 45 % to over 70 % (Mc76). When skeleton uptake is found to be lower, a correspondingly higher uptake in the liver is noted. These data are in general taken from inhalation cases, and the distribution seems to be a function of the physico-chemical form of the aerosol (Pr79).

5.1.3 Intestinal uptake of plutonium-239 in adults

A. Recent studies

At present, the gut uptake of plutonium is being studied with the aim of establishing gut uptake factors for members of the general public. The influence of different environmental and chemical factors are being taken into consideration, as well as concentration effects. Absorption in neonates and young animals has also been studied.

B. Influence of chemical form

Uptake has been studied in relation to soluble or insoluble plutonium, valence, complexing agents and incorporation into biological material.

I. **Insoluble compounds.** ICRP 30 differentiated between insoluble compounds of plutonium (oxides and hydroxides) and other compounds. A ten times smaller f-value is adopted for insoluble compounds. Harrison found in a recent survey that measured absorption fractions vary from about 0.02 % for plutonium-239 oxide in the rat to 0.000003 % for plutonium-238 oxide in the pig (Ha82a). The wide range of variation is probably a reflection of various physical and chemical characterstics of the plutonium oxide, such as particle size and specific activity. For insoluble compounds Harrison suggests no change in the ICRP f value for adults', for children, he proposes 0.1 % for the first three months and 0.05 % for the first year.

II. **Oxidation state.** Earlier, much interest was centered on the fact that plutonium might be present in the environment in the hexavalent form and that absorption of this Pu(VI) from the gastrointestinal tract might, under certain circumstances, be higher than that of Pu(IV). Environmental studies have shown that plutonium originating from the Windscale accident in solution in the Irish Sea was predominantly hexavalent (Ne78). A study by Larsen and Oldham had also shown that chlorination of drinking water would oxidize plutonium to its hexavalent state (La78). Weeks et al. reported in 1956 (We56) a thousand times higher uptake of Pu(VI) in fasting rats compared to Pu(IV).

Recently, however, Larsen et al., Stather et al. and Sullivan et al. have found that ingestion of plutonium as Pu(VI) does not enhance gut absorption in either fasting or non-fasting animals (St80, Su79b, La81a, La81b, St81). Sullivan also studied the experimental conditions used by Weeks et al. and found that the increased absorption of Pu(VI) in rats was attributable to the use of fasting animals and dichromate oxidant in combination (Su79b, Su81b). According to Harrison, it can therefore be concluded that, under normal non-fasting conditions, the absorption of plutonium is independent of

the valence state in which it is ingested, probably because the higher oxidation states are reduced to Pu(IV) in the acid conditions of the stomach (Ha82a).

C. Low concentrations and protracted administration

In general, the uptake of soluble plutonium is obstructed by hydrolysis and the formation of hydroxide polymers at physiological pH in the intestine. This leads to a higher uptake of plutonium citrate complex, due to the stability of this compound. The rate and extent of polymer formation increase as the concentration of the solution is increased. Protracted administration of a given mass of plutonium can therefore be expected to limit polymer formation within the gastrointestinal tract, and increase the proportion available for absorption in soluble form (St79).

Stather et al. have investigated plutonium uptake in hamsters after ingestion of drinking water containing 0.2 pg/ml of plutonium-236 (St81). With this concentration of Pu-239 in drinking water, the annual intake would amount to 0.2 % of the ICRP 30 limit for workers. Both tetravalent and hexavalent plutonium in solution were administered, as well as plutonium citrate. The observed absorption ranges from 0.001 % to 0.003 %. This result indicates that ingestion of very small quantities of plutonium in drinking water will not lead to absorption levels above the 0.01 % adopted by the ICRP for soluble compounds of plutonium. The low value obtained for plutonium citrate in this experiment is explained by the authors by saying that at these low concentrations, the citrate used was insufficient to prevent hydrolysis. In an earlier report by Katz et al., an absorption of about 0.003 % was observed when rats were fed chronically for 9.5 months with plutonium-238 nitrate in a concentration comparable to the M.P.C. (earlier ICRP recommendation of maximum permissible concentration (ICRP59)) (Ka55).

Sullivan, however, reports the opposite result for mice (Su81c). A group of adult mice received plutonium-237 nitrate in 0.15 ml at a concentration of 27 pg/ml in a single dose. Another group received plutonium-239 at a concentration about one million times higher. The absorption of plutonium-239 was 0.01 %

(equal to the ICRP 30 value for humans), and the observed absorption of Pu-237, fed at the lower mass level, was about 10-fold higher. This experiment confirms previous observations (Su80a). One possible explanation for the discrepancy, besides the difference in administration, is the different choice of species in the different cases. Hamsters are known to absorb less plutonium then mice or rats (Ha82b). Sufficient data is not yet available to make a reliable estimate of the absorption in humans.

D. Incorporation into biological material

The absorption of plutonium incorporated in biological material has been studied by feeding laboratory animals with liver from other animals injected with plutonium, or with crops that have taken up plutonium from the soil. It was early recognized that different complexing agents, such as citrate or DTPA, increased the absorption of plutonium from the gut, as well as biological availability in general (Ca47, We56, Ba72, Li76, Ba78). This effect is due to the chemical stability of the complexing agents which prevent hydrolysis. An extensive study of the influence of complexing agents present in foodstuffs has been made by Harrison et al. (Ha81b).

In animals fed with plutonium incorporated in liver, an absorption of up to 0.2 % has been observed when the feed-producer had been given nitrate (Su81a). With liver from animals to whom citrate had been given, uptakes of 0.05 % were observed by Sullivan (Su81a), while Harrison et al. observed values of 0.01 % and 0.03 % (Ha82a).

Sullivan and coworkers also performed feeding experiments on animals using alfalfa grown on soil to which plutonium nitrate had beed added (Su80c). Their experiments indicate an increased availability of the plutonium in this case, even if the increase is not by a large factor.

A much higher increase in availability has been noted when chelating agents such as DTPA have been used. Baxter and Sullivan found a 700-fold increase in gut absorption with this agent (Ba72). Even if this is compensated for to a great extent by rapid excretion through the kidneys, the net effect was to increase

plutonium retention in bone and liver by factors of 2 and 6, respectively. Recently, Harrison et al. studied the absorption of plutonium bound to different organic compounds common in food (Ha81b, Ha82a). For phytate absorption, factors of 0.1 % in rats and 0.01 % in rabbits were observed. Phytate is a complex present in a large number of foodstuffs including potatoes, cereal crops, peas and beans. They also studied different chelating agents present in the form of organic materials in soil and water. They used plutoniun-238 for these studies, and gut absorption was observed to increase by a factor of up to five compared to the absorption of plutonium citrate.

E. Effect of diet and other minor factors

In the body, plutonium has been shown to be associated with the iron-transport protein transferrin and the iron-storage protein ferritin (Ta73). Since iron absorption from the gut is regulated by the iron status of the body, the absorption of plutonium has been studied in relation to the iron content of the diet and the body and to iron metabolism. The results of these studies seem, to contradict each other however (St79,Ha81b).

Studies have also been reported of the influence of different drugs as well as of special foods, such as orange juice, on the gut absorption of plutonium.

It is well known that fasting increases the absorption of plutonium from the gut (Ha82b).

5.1.4 Metabolism and absorption in newborn and young animals

Higher values are obtained for the absorption of plutonium in newborn than in adults; typically if increases 100-fold in the newborn (Ba58, Su80b, Ha82a). This is also the case for most other actinides. The highest value observed is in one day-old swine, where Sullivan observed values of 10-80 % for animals given plutonium nitrate (Su80b). Both hamsters and rats show progressive reduction of absorption during the suckling period. By the time the animals are weaned from milk to solid food, the adult level of absorption is reached (Ba58, Ha82a). A similar result has been observed by Sullivan in swine (Su79c). The higher absorption may be a consequence of the special structure of the intestine wall

found in neonates during the period of lactation. Permeability of the intestine to proteins and other macromolecules is essential in order for the child to acquire passive immunity.

Children and adolescents also have different bone structures and bone metabolism. In general, radionuclides tend to be deposited on the metaphyseal growth complex.

5.1.5 Possible future changes

Suggestions have been made to the ICRP to increase the recommended factor of gut uptake of plutonium for use in dosimetry calculations for members of the public (No81,Th82). D M Taylor proposes a single value of 0.1 % for absorption of all compounds of plutonium, and H Smith and colleagues at the NRPB (National Radiological Protection Board) have proposed to ICRP committee 2 an f value of 0.05 % for all dietary intake by adults, with the exception of oxides, for which a value of 0.001 % is suggested (Ha82a). The reason for these suggestions is to be found in the related recent studies of absorption of biologically incorporated radionuclides, and absorption of low concentrations of radionuclides over an extended period of time.

A more sophisticated bone model that allows for burial and recycling might well be applicable to all surface-seeking radionuclides (Pr79). In the case of plutonium, the use of this model would result in a dose conversion factor to the bone surfaces of 0.61 Sv/MBq instead of 2.1 Sv/MBq, which can be calculated from the ICRP surface seeking model. This decrease of the dose factor would lead to a corresponding increase of the ALI by a factor of 3.5.

For the first year of life, it is recommended that an enhancement factor of 100 for absorption should be applied during the first three months. After that, during the weaning period, the enhancement factor should decrease linearly to 1, this value being reached at an age of nine months (Ha82a). This recommendation originates from a review of gut uptake data for 21 elements made by Thompson, where he propose that an enhancement factor of 100 should be applicable to all elements with an absorption of less than 0.1 % (No81).

5.2 Neptunium-237

Neptunium-237 is the first member of the fourth radioactive decay chain. The isotope is an alpha emitter with a particle energy of 4.77 MeV. Its physical half-life is about 2 million years, which is about 100 times that of plutonium-239. The chemical properties of neptunium are similar to the properties of uranium with regard to behaviour in solutions and to those of plutonium with regard to reactivity.

Until recently, little attention has been given to the problems of distribution and the biological effects of neptunium, and only a few references can be found with biokinetic data for neptunium-237. However, it has recently been recognized that if the risk figures of the 1980 report BEIR III (NAS80) and the metabolic models of ICRP 30 are combined, Np-237 is found to dominate the hazard from buried high-level radioactive waste at a time around one million years after deposition (Co82).

A comprehensive review of the current biological and environmental literature on neptunium performed by R C Thompson, also a memeber of ICRP committee 2, has recently been published (To82). This review also critically examine the ICRP 30 model for neptunium.

5.2.1 The ICRP 30 metabolic model

With the publication of ICRP 30 (ICRP80), the recommended f-value was raised by a factor 100 from the earlier recommendation in publication 2 (ICRP59). The uptake value now used by the ICRP is 1 % for all compounds of neptunium (ICRP80). The ICRP base their f value on results from rat experiments performed by Ballou (Ba62) and Sullivan (Su75, Su76). For neptunium nitrate or citrate, Ballou observed gastrointestinal absorption factors of between 0.12 and 2.3 % (Ba62), depending on chemical and physical state. The most readily absorbed neptunium was in the hexavalent state. Sullivan found absorption factors of 0.5 % for male rats and 1.2 % and 1.5 % for female (Su76, Su75). ICRP 30 also includes the remark that the degree of absorption of neptunium in trace quantities, or incorporated in food, may be a factor of ten lower (Ba62, Su76).

Concerning the distribution and retention of neptunium, ICRP 30 has used the same metabolic model as for plutonium, as experimental data indicate that the metabolic behaviour of those two elements is similar (Ba62, Su75, Su76). Thus, 45 % is assumed to be translocated to the mineral bone and 45 % to the liver. The ICRP also uses a fractional translocation to the gonadal tissues of 0.00001 per gram, which means a seven-fold higher concentration compared to a homogeneous distribution. The figure originates from a literature review by Richmond and Thomas (Ri75), and is about 10 per cent higher than their result. Neptunium not translocated to bone liver or gonads is assumed to go directly to excreta. Its biological half-life in mineral bone is 100 years, in the liver 40 years. Neptunium deposited in the gonads is assumed to be retained there indefinitely.

For neptunium-237, annual intake is limited by the absorbed dose equivalent to bone surfaces.

5.2.2 Intestinal uptake of neptunium-237

Since the publication of ICRP 30 (vol II), studies of gastrointestinal absorption of actinide elements including neptunium in adult and neonatal rats, guinea pigs and dogs have been presented by Sullivan (Su80a, Su80b). Harrison and Stather have studied the absorption of neptunium in the adult hamster (Ha81a). The results of the different studies are presented in table 5.1.

Sullivan administred pentavalent neptunium as a soluble nitrate compound orally to rats. Neptunium absorption was found to be the highest among the different elements investigated (Th, U, Np, Pu, Am, Cm, Cf and Es). 1.2 % was recovered in the skeleton, liver or urine, (0.73 % in the skeleton and liver only). This in spite of the relatively high mass administred (Su80a). The author attributes the high absorption of neptunium compared to the other elements to its stable pentavalent oxidation state.

Table 5.1. Gut uptake of neptunium.

Chemical form	Laboratory animal	No of animals	Absorption %	Rem	Reference
Citrate	Hamster	6	0.062	0.5 mg	Harrison,82
Nitrate		12	0.052	-"-	-"-
Nitrate	Rat	6	0.26	-"-	-"-
Nitrate	Rat	6	0.063	5 pg	-"-
Nitrate	Rat F	15	1.15		Sullivan,76
Biologically incorporated	Rat F	15	0.03	Skeleton and liver	-"-
Nitrate	Rat M	7	0.56	-"-	-"-
Biologically incorporated	Rat M	7	0.05	-"-	-"-
Nitrate	Rat 2d	5	0.8	lower than stated dose to the skeleton	-"-
	4d	6	0.5		
	8d	10	1.2		
Nitrate	Rat	11	1.5		Sullivan,75
	Rat 8d	10	6.6		-"-
Nitrate	Rat	14	1.2	8.5 mg	Sullivan,80
	Rat 1d	3	0.75	2.8 mg	-"-
	2d	4	0.60	-"-	-"-
	3d	9	0.37	-"-	-"-
	4d	3	0.64	-"-	-"-
	9d	10	1.2	-"-	-"-
Industrial dust	Rat	6	0.9		Ballou,62
-"-		3	0.1	Chronic intake for 1 month	-"-
Nitrate		6-9	0.12		-"-
Citrate		6-9	0.26		-"-
Tetravalent		6-9	0.30		-"-
Pentavalent		6-9	1.1		-"-
Hexavalent		6-9	2.3		-"-

Harrison and Stather used neptunium as a citrate and in nitrate form. They studied both gut absorption and the distribution of neptunium in citrate form after intraveneous injection. The values obtained for total absorption in the gut were 0.05 % and 0.06 % (no significant difference) for the nitrate and the citrate complex, respectively. These figures are low compared to other published results for uptake. One factor that may have contributed to the difference is that the adminsitered mass of neptunium was lower, 0.5 mg, in this case, and another factor that hamster was used as experimental animal. To study the influence of the mass, the uptake of neptunium-239 (T 1/2 = 2.35 d) was studied in rats by Harrison et al. (Ha82b). The administred mass was only 5 pg of neptunium, and the total uptake measured was 0.063 %. In a control group of rats receiving 0.5 mg of Np-237, an uptake of 0.26 % was measured.

Absorption data were usually observed to be about 100 times higher in the neonatal animal than in the adult rodent (Su80b). For neptunium, however, the observed increase was found to be less than a factor of 10. In this case as well the unique behaviour of neptunium in soluble form, is attributed by the authors to its stable pentavalent state. The stability of the oxidation state reduces the probability of the formation of polymeric hydroxides that might be absorbed more efficiently by the neonate.

Ballou and coworkers (Ba62) have also studied the gastrointestinal absorption of neptunium in the form of industrial dust in rats given a single dose. The absorption was found to be 0.9%. If the suspension of dust was fed chronically to the animals for one month, only 0.1% of the administred dose was retained at the end of the period.

Sullivan has also studied retention in the intestinal contents and wall in rats 2 - 9 days old (Su80b). For a gavaged dose of 2.8 mg neptunium nitrate, up to 20 % was found in the wall of the intestine 7 days after administration.

5.2.3 Reliability of the ICRP 30 model

The specific activity of neptunium-237 is about 26 MBq/g, and as it is the most long lived isotope of neptunium, the mass for the ALI of neptunium-237 (3 kBq)

can be decided to 0.1 mg. With this low intake mass gastro-intestinal uptake seems to be lower (on the order of one tenth) than the value recommended by ICRP 30. Of special interest is the decrease in uptake of neptunium bound to animal tissue which has been observed (Su76).

Based on the available results found in the literature at that time, the f-factor chosen by ICRP 30 seems to be a reasonable pessimistic estimate. Further investigations are needed to be able to make a reasonable estimate of the absorption of neptunium under the special conditions that exist for neptunium present in effluents from a leaking underground nuclear waste repository. Some results indicate that studies like this will show that the f factor could be lowered by about a factor of ten for neptunium in low concentrations in the environment. According to Thompson, this is also a development in the foreseeable future that might change the ALI by approximately 10 (To82).

Nenot and coworkers (Ne72) have also found some indications that neptunium in bone is more similar to calcium than plutonium. This means that the element would be incorporated in mineral bone to some degree, and thus that the irradiation of the sensitive parts of the skeleton, endosteal surfaces and red marrow, would decrease. This would cause the ALI value to rise.

In his review, Thompson recommends, in contrast to ICRP 30, that the initial activity ratio between bone and liver should be changed from unity to 4, 60 % uptake in bone and 15 % in the liver, the remaining 25 % being quickly excreted (To82). The ICRP retention times are kept. This has the effect that the dose to bone surfaces increases, while the dose to the liver decreases. The resulting decrease in ALI will be small, only about 15 %. Even though it is not a recommendation of the author, the increase in the dose to the bone surfaces is compensated for by, the assumption that the distribution of neptunium in the skeleton more closely resembles a volume distribution than a surface distribution. A more reliable demonstration that this is the case will probably increase the ALI by a factor of five in the future.

5.3 Radium-226

Radium-226 is a natural by occuring isotope, included in the uranium-238 decay chain. It has a physical half-life of 1600 years. The content of radium-226 in soil, biota, animals and man in different parts of the world is fairly well known. These data are compiled in UNSCEAR (UN77).

There is a group of several thousand humans who acquired internal burdens of radium by ingestion, injection or inhalation some 50-60 years ago. They comprise a unique and irreplaceable human study group of the utmost importance. Particularly in the important field of dose-response relationships for humans in the very low dose domain.

5.3.1 The ICRP 30 metabolic model

The ICRP has taken f to be 20 % for all compounds of radium. As a basis for this, they refer to their publication 20 (ICRP73). They cite values of fractional absorption in the range of 0.15-0.21, derived from studies in which radium was present in drinking water, or incorporated in food.

A quite comprehensive model for the retention of radium in adults has been described in the ICRP report on alakline earth metabolism, publication 20 (ICRP73). With this model, is it possible to calculate the total number of nuclear transformations in different tissues of the body. This publication, however, focuses heavily on metabolic response to transient intake, because of the importance of occupational and medical radium cases.

Ra-226 decays to Rn-222, which has a radioactive half-life of 3.8 days. Since it is a gas, any Rn-222 present might be assumed to escape from the body without decaying. The fraction of Ra-222 reatined in mineral bone depends on the depth of deposition of the parent nuclide, which is a function of the time it has been resident in the tissue. As an average the ICRP assumes that 30 % is retained on the site of formation, as estimated from Norris et al. (No55).

The annual intake of radium-226 is limited by the absorbed dose equivalent to the bone surfaces.

5.3.2 Gastrointestinal uptake of Ra-226

Experiments with young rats, performed by Taylor and coworkers, have shown a higher uptake of $RaCl_2$ from the gastro-intestinal tract than in adult rats (Ta62). These investigators found that uptake was as high as 79 % in 14-18-day-old rats, compared to 3.2 % found in 60-70-week-old animals. This study has also shown that starvation increases uptake by about 50 % for adult rats. The authors explain this with reference to the presence of sulphate ions in the food, which might be expected to reduce radium to insoluble radium sulphate. They fail, however, to confirm their theory experimentally.

In 1969, Maletskos and coworkers published results from measurements in man of the absorption of radium-224 (T1/2 = 1.8 h) in sulphate form (Mal69). From five normal subjects, they obtained an average absorption factor of 0.20. This should be the fraction at which radium was absorbed in the humans that were exposed 30-40 years ago via dial painting. Against this background, the ICRP uptake value of 0.2 seems to be a reasonable estimate, and it is improbable that this figure will be appreciably changed in the future.

5.3.3 Deposition of Ra-226 in the body

The high natural background radiation that can be found at certain locations in the word, for example Kerala in India, offer a unique opportunity to study the deposition and metabolism of radium in man under conditions of chronic exposure. The important route of intake for Ra in these areas is via food and water (Wr77). In order to estimate the radium concentration in the human body on the basis of dietary information, calcium intake should be known as well. The Kerala body burden is about 40 % greater than the burden in the United States (Wr77).

When radium is absorbed into the body, it behaves chemically like calcium. Under chronic exposure conditions, the ICRP model for alkaline earth suggests that 85 % of the total body burden of radium is found in the skeleton (ICRP73, Wr77). It is initially deposited on bone surfaces and in areas of active bone turnover. The initial surface deposit gradually migrates to produce a diffuse distribution throughout the entire bone mineral (ICRP73). 15 % is deposited in

soft tissue (ICRP73,Wr77). In general, the pattern for radium deposition in the adult human skeleton is that of an overall diffuse distribution with superimposed hot spots (ICRP68). The hot spots are now known to correspond to radium deposition along individual Haversian systems which were forming at the time of intake (Ev74).

5.4 Thorium-230

Thorium-230 is as a daughter in the uranium-238 radioactive decay chain, a naturally occuring radioisotope. It is an alpha-decaying radioisotope, and has a half-life of 80000 years.

Thorium has many chemical and biological properties in common with plutonium with regard to biokinetics and transport in the environment (Wr77), although the specific activity of thorium is much less than that of plutonium. Studying of the distribution of thorium will thus yield certain information on plutonium, and vice versa. Extensive studies have been made of certain insoluble compounds of thorium, especially thorium-232 in the form of Thorotrast, which has been administred for radiodiagnostic purposes. Thorium in soluble form, however, has received comparatively little attention.

The concentration of natural thorium (mainly Th-232) in the human body has, unfortunately, not been adequatly studied, probably because the absorbed dose is small compared to that of other natural radionuclides (Wr77).

5.4.1 The ICRP 30 Metabolic model

Most literature shows that thorium is highly discriminated against in the gastrointestinal tract. ICRP 30 has chosen an f-falue of 0.02 % for all compounds of thorium (ICRP79). This figure is based mainly on studies on humans by Maletskos and coworkers (Mal69). Experiments on the absorption of Th-232 as a nitrate complex from the gastrointestinal tract of the rat by Traikovich (Tr70) indicate, however, an uptake value of about 0.5 - 1 %.

Of the thorium entering the body, ICRP 30 assumes that 70 % is deposited on bone surfaces, 4 % in the liver, 16 % is homogeneously distributed in the rest of

the body and 10 % is promptly excreted. In bone, the biological half-life of thorium is 8000 days, and in soft tissue 700 days. This model has been taken from Stover and coworkers (St60, St65). However, in a study published in an annual report by the Pacific Northwest Laboratories, Sullivan and Wogman administered Th-228 nitrate intragastrically to rats (Su79). They noted, unexpectedly, that 80 % of the thorium absorbed through the gut was retained in the muscle. The authors make no attempt to explain this deviation from earlier studies. One reason may be that thorium builds up in the different organs relatively slowly. Kendysh (Ke66) has found that the thorium level in the liver reaches its maximum 90 days after oral administration, and that the skeletal content is still increasing after 360 days. The increase in the skeleton is, however, only a factor of 2.5 between the first and the 360:th day.

The annual intake of thorium-232 is limited by the absorbed dose equivalent to the bone surfaces.

5.4.2 Reliability of the ICRP 30 model

The gut uptake of thorium seems to be comparatively well estimated. Thorium in nitrate form can exist in a soluble state only at pH < 3.5-3.6. At higher pH, which can be caused by contact with biological tissue, hydrolysis will occur and insoluble thorium will be formed. The relatively high uptake noted by Traikoivich could be explained by supposing that thorium was not completely hydrolysed. It is improbable that the same high uptake figures could be obtained in man.

Sullivan and Wogman have studied the gastrointestinal absorption of Th-228 nitrate at pH 2, administred to newborn and adult rats (Su79). Adult rats were found to retain 0.025 % and neonates 1.2 % of the thorium administred, which is close to the ICRP value.

Ra-226 and its daughter Rn-222 (T 1/2 = 3.8 d) are components of the decay chain that starts with Th-230. The assumption that the daughter stays on the site where it was created overestimates the dose to the bone surfaces in this case, since radium presumably diffuses into the volume of the bone. The ICRP

also calculates the ALI on the assumption that no radon escapes from the bone. Owing to the long half-life of Ra-226, the contribution made by the daughters of Th-230 to the total absorbed dose will only amount to about 5 % of the total dose. These assumptions are therefore only small approximations. The escape of 30 % of the created radon would reduce the absorbed dose by 3 %.

5.5 Protactinium-231

Protactinium is one of the rarest of the naturally-occuring elements. It is also one of the least studied elements with respect to metabolism in humans and animals. Very few references are found in the litterature. Protactinium-231 has a physical half-life of 32 800 years.

5.5.1 The ICRP 30 metabolic model

The ICRP has adopted an f factor of 0.1 % for all compounds of protactinium. This factor is based mainly on experiments with rats by Hamilton 1948 (Ha48), and on later experiments, also with rats, by Zalikin (Za66a, Za66b, Za69). These experiments have given results of between 0.006 % and 0.2 %.

Retention and distribution measurements have been performed on a man accidentially contaminated with Pa-231 through a puncture wound in the hand (Ne68, Ne74). Based on this study, and on the above-mentioned studies by Hamilton and Zalikin, the ICRP assumes that 40 % of the protactinium absorbed from the gut is translocated to the skeleton, 15 % to the liver and 2 % to the kidneys. The remaining fraction goes directly to excretion. By analogy with plutonium and transplutonic actinides, the biological half-life of protactinium-231 in the skeleton is assumed to be 100 years. Protactinium deposited in the liver or in the kidneys has a biphasic retention (Za69), with half-lives of 10 and 60 days. In the liver, the relative fractions between these half-lives are 0.7 and 0.3, respectively, and in the kidneys 0.2 and 0.8, respectively.

The annual intake of protactinium-231 is limited by the absorbed dose equivalent to the bone surfaces.

5.5.2 Gastro-intestinal uptake of Pa-231

Since the publication of the ICRP 30, gut uptake of protactinium has been studied by Harrison and Stather in England (Ha81a). Through gastric intubation, they administred Pa flouride and Pa as a citrate complex to hamsters. They obtained an absorption of 0.22 % for the fluoride, while the absorption of Pa bound to the citrate complex was found to be as high as 3.9 %. They suggest that the ICRP estimate of 0.1 % absorption may be low, and believe the value of 1.0 % for all compounds of protactinium to be a more accurate one. The authors gives no other plausible explanation for the difference between their results and the results of the work cited by the ICRP 30 then that they used hamsters instead of rats. They also emphasize that one of the earlier reports says "the uptake does not exceed 1 - 2 % and may be as low as 0.006 %" (Za 66a, Za66b). The possible range of variation of the result is thus quite large.

5.5.3 Reliability of the ICRP 30 model

The distribution and retention of protactinium seems to be estimated with a higher degree of certainty than gut uptake. The half-life of 100 years is, as for plutonium, a conservative estimate. The experimental evidence for this is a recorded biological half-life in excess of 100 days. The assumption, in analogy with other actinides, that protactinium is a surface seeker is also a conservative one, since no experimental data concerning distribution in the skeleton can be found. As for plutonium and neptunium, it is be possible that this assumption overestimates the absorbed dose in the bone surfaces by about a factor of 5.

In the light of the only recently published report on the absorption of protactinium (Ha81a), the uptake factor of 0.001 recommened by the ICRP seems to be low. In England, the NRPB (National Radiological Protection Board) has used an f value of 1 % for calculating ALI in their report NRPB-R82 (Ad78). This value was adopted by analogy, not stating with which element. Thus, for protactinium, they use an f value that is 10 times higher than the ICRP's. The NRPB report is based largely on the same data as the ICRP 30, and was published in advance for many radionuclides, including protactinium. The ICRP changed to a lower value at a late stage in the preparation of their report

(Th82). The situation is unclear, and further research is required, especially on protactinium bound to biological material, for which, in analogy with plutonium, a higher gastro-intestinal uptake may not be unexpected.

5.6 Technetium-99

Technetium-99 is a long-lived fission product ($T_{1/2}$ = 21 2000 years). It is a pure beta-emitter, with a maximal beta energy of 292 keV.

No stable isotope of technetium exists. Sixteen isotopes of technetium are known, with atomic masses ranging from 92 to 107. The most long-lived isotope, Tc-97, has a half life of 2.6 million years, which is too short for the element to be found in nature. The behaviour of technetium in the environment is therefore not well known.

5.6.1 The ICRP 30 metabolic model

Due to its favourable physical properties, technetium-99m has been wideley used in medicine. It is therefore well known that technetium in ionic form is absorbed from the gastrointestinal tract to a relatively large extent. However, this isotope has a half-life of only six hours, so its metabolism and long-term retention have not been very well studied.

The ICRP has adopted an uptake factor of 0.8 for all compounds of technetium. Although higher factors have been reported (Be66), it has been shown on humans that technetium administered as pertechnetate is erratically absorbed with marked variability as to the time and extent of absorption (Ha73). Technetium chloride in rats has been found to be absorbed to a lesser degree, 50 % (Su78b).

Technetium that has entered into the blood is distributed between the thyroid (4 %), the stomach wall (10 %), the liver (3 %) and the remaining tissues (83 %). The biological half-lives are, according to the ICRP, relatively short, the longest being 22 days (5 %).

The annual intake of technetium-99 is limited by the effective dose equivalent, the stochastic effect.

5.6.2 Reliability of the ICRP 30 model

Since the gastro-intestinal uptake, in this case, is relatively large and well known, uncertainties in this parameter will not be a major source of error in the total dose conversion factor. The long half-life, however, introduces another source of error. Although it is impropable, a small undetected component might be retained in the body for a long period. The most likely site of such a component would be in the skeleton. The factor by which the dose conversion factor is increased due to the presence of a long-term component is illustrated in figure 5.1. The influence of the ALI of this hypothetical component is illustrated in figure 5.2.

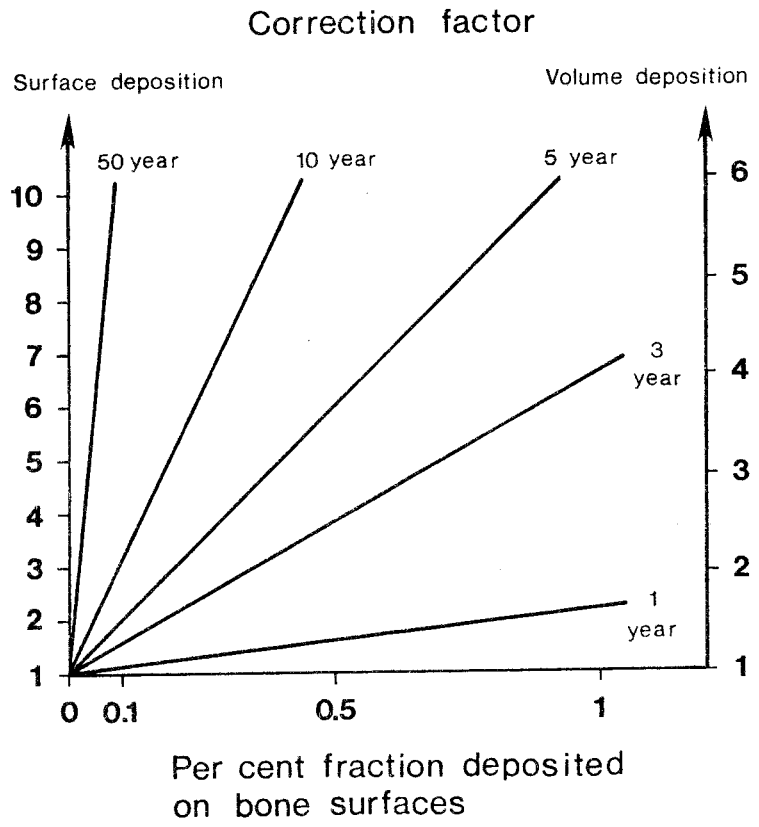

Figure 5.1. Influence on effective dose equivalent from ^{99}Tc of a long-term retention component in the skeleton.

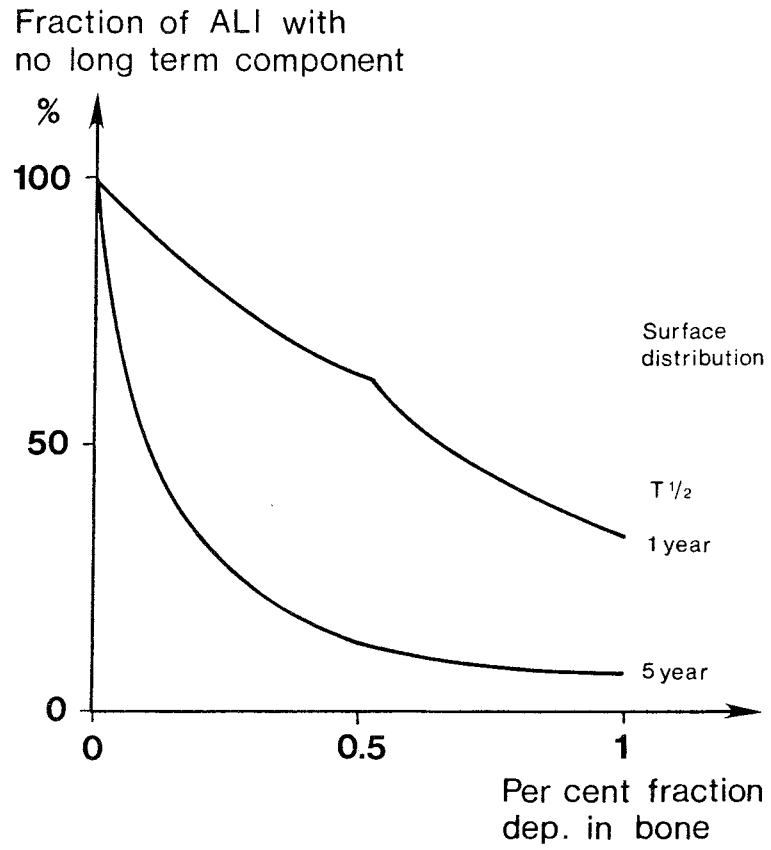

Figure 5.2. Influence on ALI for ^{99}Tc of a long-term retention component on bone surface.

5.6.3 Gastro-intestinal absorption

Very few reports dealing with the effect of biological incorporation of technetium on absorption have been found. Sullivan and coworkers used Tc-95m in rats and guinea pigs (Su78b). They found that the absorption of technetium incorporated into either animal or plant tissue was about half of that of inorganic pertechnetate. The elimination rate was not altered by the incorporation. This study also included newborn rats. They were found to have a slower elimination rate than adults, up to two weeks post administration. Most of this retained activity was found in the pelt. Recent studies indicate that the pertechnetate ion is an analog to the sulfate ion (Ga82). The fraction transferred to eggs has therefore been measured in birds fed with ionic or plant-incorporated technetium. Birds given plant incorporated technetium showed less transfer to eggs.

Experimental data are too few to draw any conclusions concerning gut absorption and metabolism of technetium in adults.

5.7 Iodine-129

Iodine-129 is a long lived iodine radioisotope with a physical half-life of 16 million years. It decays to Xenon-129 via beta-minus. In its decay, several low-energy Auger electrons with energies of up to 4 keV (several with energies less than 1 keV) are emitted. The range of these electrons is less than 100 nm.

5.7.1 The radiotoxicity of iodine-129

Due to the presence of low energy electrons, with ranges less than the size of most cells, cellular damage due to radiation will depend largely on intracellular radionuclide distribution. Iodine in a form that can be incorporated into the DNA of the mammalian cells is extremely radiotoxic. In contrast, iodine attached to the outer membrane of the cell is surprisingly nontoxic. This high toxicity of iodine incorporated into the cell nucleus is caused by the dense shower of low-energy electrons, which results in a highly charged daughter atom and a high density of electron irradiation in the immediate vicinity of the disintegrating radionuclide (Bl78,Ho81).

The way that iodine could be incorporated into the cell nucleus is via thyroid and triiodothyronine, T3. Certain cells can be found in different organs throughout the body, mainly in the brain, whose nuclei contain receptors for T3, especially in the hypophysis, and particularly in children. The number of receptors is limited, however, and when all sites are occupied, no more T3 can be taken up in the nuclei. From this limitation it follows that the decays of a high-specific-activity radionuclide will more frequently take place within the cell nuclei than the decays of a low-specific-activity radionuclide. The specific activity of iodine-129 is so low (6.5 Mbq/g) that its activty within the cell nuclei will be almost neglible (Jo82).

5.7.2 The ICRP 30 metabolic model

A vast amount of literature exists concerning metabolism of more short-lived isotopes of iodine, such as I-131 and I-125, which are of importance both in

medicine and health physics. There is also a stable isotope of iodine, I-127, which can be used in biological studies.

Iodine is absorbed rapidly and almost completely from the gastrointestinal tract (ICRP79). The ICRP has therefore taken f to be unity.

The metabolic model of iodine is in essence that of Riggs et al. (Ri52). It is a simple three compartment model, outlined in the figure below.

30 % of the iodine is assumed to be translocated to the thyroid, where it is retained with a biological half-life of 120 days. It is cleared from the thyroid in the form of organic iodine. The compartment for organic iodine, as well as the transfer compartment, are assumed to be homogeneously distributed throughout the body.

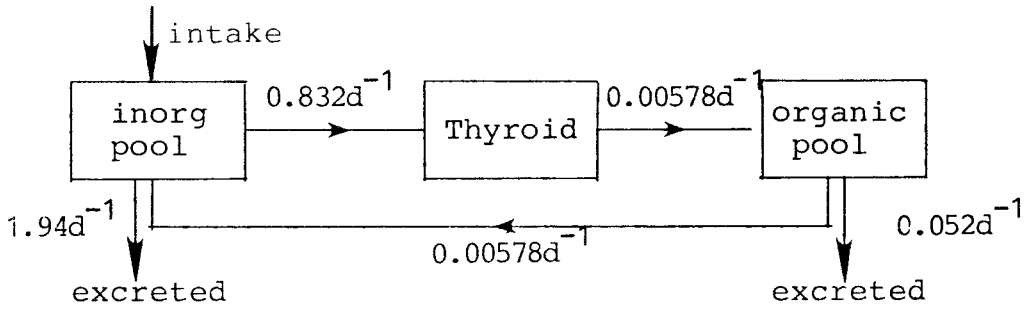

Figure 5.3. ICRP 30 model for iodine.

With recirculation of iodine, its real half-life in the thyroid will be longer than 120 days. The ICRP has, however, not taken this into account in their calculation of ALI. Their dose factor for the thyroid (which receives a completely dominating absorbed dose to a single organ in this case) is underestimated by 25 %.

The annual intake of iodine-129 is limited by the absorbed dose equivalent to the thyroid.

5.7.3 Reliability of ICRP 30 model

The uptake of iodine in the thyroid gland is controlled by a number of factors including the thyroid functional status of the individual, the size of the body iodine pool and the quantity of iodine in the diet. For any population with normal thyroid function, the mean uptake of iodine may vary with geographical area due to variations in the iodine content of the food and water. For most normal populations, the upake of radioiodine appears to lie in the range of about 10 to 40 % (Ta81). A more extensive statistical study of the variability of iodine uptake was presented by Dunning and Schwarz in 1981 (Du81). The value of 30 % seems to be rather close to what, at present, is considered normal in Sweden (No81). The uptake of iodine-129 would, however, be expected to be smaller, due to its low specific activity. One ALI, 0.2 MBq, corresponds to about 30 mg iodine; a sufficient iodine intake is 150-200 μg daily (Ka80), and this is less than one per cent of the ALI. Ingestion of only one tenth of an ALI would therefore significantly increase the normal systemic extrathyroidal iodine pool, and so slightly reduce thyroidal uptake (Ka80). It is remarkable that a sufficient iodine intake met solely by iodine-129 will not exceed the ALI by more than a factor of two to three.

The half-life of iodine in the thyroid is also subject to wide individual variation. It is not well known, mainly because the most interesting isotope is I-131, and it is too short-lived (T 1/2 = 8.03 days) compared to the biological half-life in the thyroid, so that accurate measurements are difficult to perform. Studies of the clearance of iodine from the thyroid have been published by Widman et al. (Wi80), who used I-125 ($T_{1/2}$ = 60 d) in search of a possible small long-term component of the retention curve. In 21 normal volunteers, they observed biological half-lives of between 61 and 251 days, with a mean of 113 days. No other compartment with a biological half-life greater than this was found.

Due to its long physical half-life, the absorbed dose from iodine-129 should be strongly influenced by a small long-term retention component. The observation of Widman et al. is therefore very important in this case. Other reports of long-term retention studies are sparse; Bordell and Wechsler reports a biological half-life ranging from 104 to 180 days, with a mean value of 130 days for five

normal individuals (Bo72). It should be noted that these data are based on relatively recent American investigations, where the normal thyroid uptake is about half of the Swedish one.

Extensive metabolic models for iodine have been published (Be72, Jo81, Ta81). The most detailed model is that of Berman (Be72), who quotes a normal biological half-life for iodine in the thyroid in the range 75-94 days for the USA population. Based on biological data from "Reference man" (ICRP75), Johnson has devised a metabolic model similar to that of ICRP 30 for both adults and children (Jo81). From his biological data, an uptake of 25 % in the thyroid can be derived, and a half-life of 128 days for the adult. This results in an absorbed dose that is about 15 % lower than that calculated from the ICRP model.

The recirculation of iodine according to the metabolic models has not been taken into account in the calculation of the ALI for I-129 in ICRP30. The correct dose conversion factor should be about 30 % higher, which leads to a reduction of the ALI by about 25 %. However, the influence of the low specific activity, and the uncertainty of the uptake and excretion parameters, make this a minor factor.

5.7.4 Iodine metabolism in children

Due to the relatively short biological half-life of I-129, in comparison with the length of childhood, the absorbed dose to the thyroid will be relatively age-dependent.

Not much is known about iodine metabolism in children. Johnson has included models for children in his review. About the same uptake, and a half-life which is about half that of the adult, can be derived from his model for a newborn child (Jo81). Taking into account the fact that the mass of a child's thyroid is about one tenth that of an adult's an effective dose equivalent that is about a factor of five greater than for the adult can be calculated.

According to Kaul and Roedler, who cite a German report, the biological half-life of iodine in a newborn child's thyroid is only about two weeks, and in a

six-month-old child just over three weeks (Ka80). They also report uptake values for children: 0-0.2 years 50 %, 0.2-0.6 years 40 % and over 0.6 years 35 %. The effective dose equivalent will be of the same order of magnitude with these data as well. In the case of the low-specific-activity radioisotope iodine-129, it should also be noted that the intake of a certain mass of iodine will have a greater effect on a child's small thyroid than on the adult's. If it is assumed that all of the iodine in the thyroid is I-129, the absorbed dose rate to the thyroid of an adult would be 4 times as great as that of a child (So76).

Dunning and Schwarz report a mean uptake of 47 % for newborn children, as well as for adolescents (6-16 years) (Du81). The figure is subject to great variation. Among the 67 newborn children, the observed range was between 6 % and 97 %, and among the 114 adolescents between 17 % and 88 %. The data were collected from earlier studies published by other authors. According to Dunning and Schwarz, the range of halv-lives reported in different studies is 4-40 days for infants. The amount of data is very limited, however. A slightly shorter half-life can be noted for adolscents than for adults.

6 Summary

The gastro-intestinal uptake factors recommended in this report are displayed in table 6.1, together with those of ICRP 30 and ICRP 2. The values deviate from ICRP 30 for the following radionuclides.

- Pu-239; the factor is based on recent studies by Harrison et al. (Ha81b, Ha82a) as well as on proposals submitted to the ICRP for increasing the value (No81).

- Np-237; the reduction of the value recommended by ICRP 30 stems from recent results obtained by at least two different laboratories which show a low gastro-intestinel uptake when the specific activity is very low (Ha81a, Ha82b, Th82a).

- Pa-231; the recommended figure has been somewhat arbitrarily chosen. It is based mainly on a study by Harrison et al. (Ha81a) in which the authors recommend a factor of 1.0 %. An increase by a factor of ten was considered too much, and was limited to a factor of five. Since there is a serious lack of gut uptake data for Pa-231, it is strange that ICRP committee 2 did not take this work into consideration.

The ICRP metabolic models were not changed in any case. Regarding neptunium, a proposal has been made by Thompson to change the distribution between the liver and skeleton (Th82a). This change would have no significant effect on the resulting dose conversion factors.

The resulting dose conversion factors are shown in table 6.2.

For children less than one year old, the uptake value should be increased by a factor 100 for plutonium, neptunium, thorium, and protactinium. For iodine-129 and technetium-99, the committed dose equivalent may roughly be estimated to be greater by a factor of 10 for infants. This factor decreases with age. Uncertainties and possible reasons for future changes are summarized in tables 6.3 and 6.4.

Table 6.1. Recommended gastro-intestinal uptake factors for members of the public, compared to the values in ICRP 30 and ICRP 2.

	This report, members of the public	ICRP 30	ICRP 2
Pu-239 (soluble)	$5 \cdot 10^{-4}$	10^{-4}	$3 \cdot 10^{-5}$
Pu-239 (insoluble)	10^{-5}	10^{-5}	$3 \cdot 10^{-5}$
Np-237	10^{-3}	10^{-2}	$< 10^{-4}$
Ra-226	$2 \cdot 10^{-1}$	$2 \cdot 10^{-1}$	$3 \cdot 10^{-1}$
Th-230	$2 \cdot 10^{-4}$	$2 \cdot 10^{-4}$	$< 10^{-4}$
Pa-231	$5 \cdot 10^{-3}$	10^{-3}	$< 10^{-4}$
Tc-99	$8 \cdot 10^{-1}$	$8 \cdot 10^{-1}$	$5 \cdot 10^{-1}$
I-129	1.0	1.0	1.0

Table 6.2. Committed Dose Equivalent to the limiting organ, and Effective Dose Equivalent per intake of unit activity (Sv/Bq). The values for this report are integrated for 70 years, and the ICRP 30-values for 50 years.

	This report, members of the public		ICRP 30	
	Limiting organ	Effective dose equiv.	Limiting organ	Effective dose equiv.
Pu-239 (soluble)	$1.4 \cdot 10^{-5}$	$7.0 \cdot 10^{-7}$	$2.1 \cdot 10^{-6}$ (bone surf)	$1.2 \cdot 10^{-7}$
Pu-239 (insoluble)	$2.7 \cdot 10^{-7}$ (bone surf)	$1.4 \cdot 10^{-8}$	$2.1 \cdot 10^{-7}$ (bone surf)	$1.2 \cdot 10^{-8}$
Np-237	$2.5 \cdot 10^{-5}$ (bone surf)	$1.2 \cdot 10^{-6}$	$1.9 \cdot 10^{-4}$ (bone surf)	$1.1 \cdot 10^{-5}$
Ra-226	$7.5 \cdot 10^{-6}$ (bone surf)	$3.3 \cdot 10^{-7}$	$6.8 \cdot 10^{-6}$ (bone surf)	$3.1 \cdot 10^{-7}$
Th-230	$4.1 \cdot 10^{-6}$ (bone surf)	$1.6 \cdot 10^{-7}$	$3.6 \cdot 10^{-6}$ (bone surf)	$1.5 \cdot 10^{-7}$
Pa-231	$5.5 \cdot 10^{-4}$ (bone surf)	$2.2 \cdot 10^{-5}$	$7.2 \cdot 10^{-5}$ (bone surf)	$2.9 \cdot 10^{-6}$
Tc-99	-	$3.4 \cdot 10^{-10}$	-	$3.4 \cdot 10^{-10}$
I-129	$3.3 \cdot 10^{-6}$ (thyroid)	$9.8 \cdot 10^{-8}$	$2.5 \cdot 10^{-6}$ (thyroid)	$7.4 \cdot 10^{-8}$

Table 6.3. Reasons for variation or uncertainty in the dose conversion factor and their relative significance.

Nuclide	Metabolic model, including dosimetry	GI-uptake	Chemical form, incl. incorp into biol. mat	Diet factors
Pu-239	*	***	**	?
Np-237	*	***	**	?
Ra-226	**	-	***	?
Th-230	***	**	*	?
Pa-231	*	***	**	?
Tc-99	**	-	***	?
I-129	***	-	-	**

*, **, *** large, larger, largest influence
- no significant influence
? not estimated

Table 6.4. Possible reasons for future changes of the dose conversion factor, except gastro intestinal uptake.

Pu-239	Refined bone model	$\frac{1}{4}$ x
Np-237	Refined bone model	$\frac{1}{4}$ x
	Other distribution between skeleton, liver and early excreta	1.1 x
Pa-231	Refined bone model	$\frac{1}{5}$ x
	Found to be a volume seeker	$\frac{1}{10}$ x
Th-230	Refined bone model	$\frac{1}{4}$ x
Tc-99	Long-term component found	10 x
I-129	Reduced thyroid uptake due to changed diet	$\frac{1}{2}$ x

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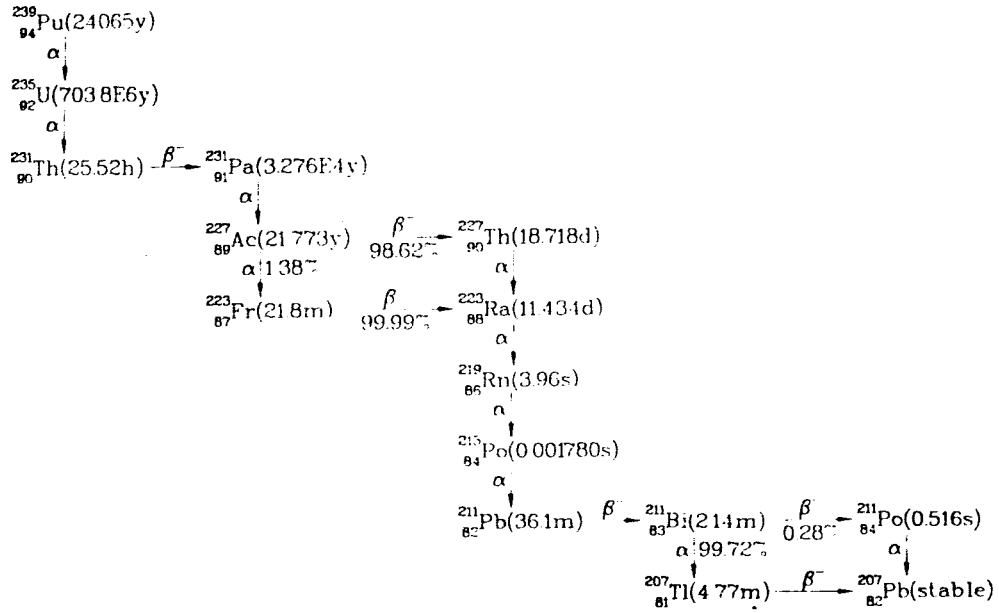
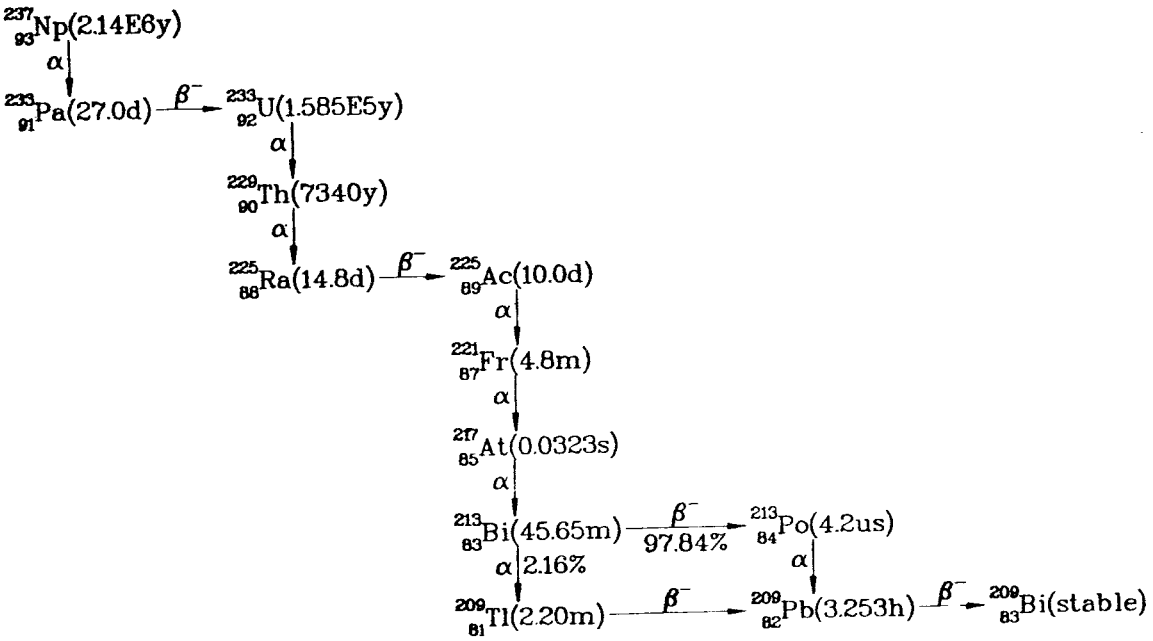
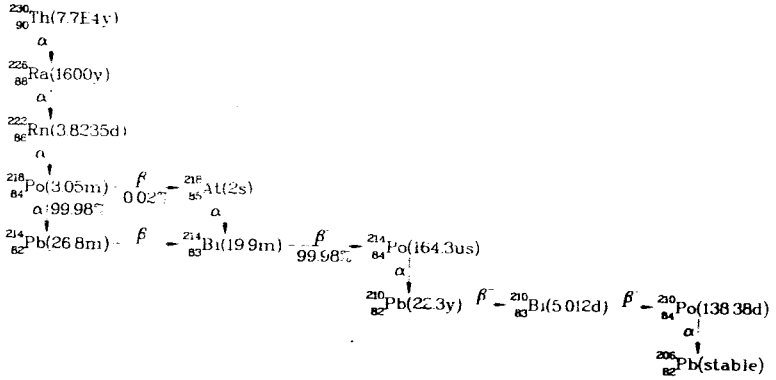
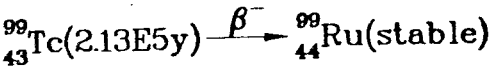
Appendix

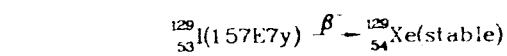
Decays chains for radionuclides included in the report.











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TR 82-01 Hydrothermal conditions around a radioactive waste repository
Part 3 - Numerical solutions for anisotropy
Roger Thunvik
Royal Institute of Technology, Stockholm, Sweden
Carol Braester
Institute of Technology, Haifa, Israel
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TR 82-02 Radiolysis of groundwater from HLW stored in copper canisters
Hilbert Christensen
Erling Bjergbakke
Studsvik Energiteknik AB, 1982-06-29

TR 82-03 Migration of radionuclides in fissured rock:
Some calculated results obtained from a model based on the concept of stratified flow and matrix diffusion
Ivars Neretnieks
Royal Institute of Technology
Department of Chemical Engineering
Stockholm, Sweden, October 1981

TR 82-04 Radionuclide chain migration in fissured rock -
The influence of matrix diffusion
Anders Rasmuson *
Akke Bengtsson **
Bertil Grundfelt **
Ivars Neretnieks *
April, 1982

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Results obtained from a model based on the concepts of hydrodynamic dispersion and matrix diffusion
Anders Rasmuson
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Royal Institute of Technology
Department of Chemical Engineering
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Calculation of rock permeability
Carol Braester
Israel Institute of Technology, Haifa, Israel
Roger Thunvik
Royal Institute of Technology
Stockholm, Sweden, June 1982

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Roland Pusch
Division Soil Mechanics, University of Luleå
Luleå, Sweden, 1982-06-30

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Part 1
Lars Birgersson
Ivars Neretnieks
Royal Institute of Technology
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PLAN 82
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PLAN 82
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TR 82-10 The hydraulic properties of fracture zones and tracer tests with non-reactive elements in Studsvik
Carl-Erik Klockars
Ove Persson
Geological Survey of Sweden, Uppsala
Ove Landström, Studsvik Energiteknik, Nyköping
Sweden, April 1982

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Klas Lundgren
ASEA-ATOM, Västerås, Sweden, 1982-08-11

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Kristina Skagius
Ivars Neretnieks
Royal Institute of Technology
Department of Chemical Engineering
Stockholm, Sweden, 1982-03-01

TR 82-13 Variation in radioactivity, uranium and radium-226 contents in three radioactive springs and along their out-flows, northern Sweden
John Ek
Sverker Evans
Lennart Ljungqvist
Studsvik Energiteknik AB
Nyköping, Sweden, 1982-06-03

TR 82-14 Oral intake of radionuclides in the population
A review of biological factors of relevance for assessment of absorbed dose at long term waste storage
Lennart Johansson
National Defence Research Institute, Dept. 4
Umeå, Sweden, October 1982