

Assessing the Reliability of Dose Coefficients for Ingestion and Inhalation of Radionuclides by Members of the Public

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ABSTRACT

Consideration of uncertainties on doses can provide numerical estimates of the reliability of the protection quantities (dose coefficients) used in radiation protection to assess exposures to radionuclides that enter the body by ingestion or inhalation ("Internal Emitters"). The ICRP system of radiological protection is reviewed and the meaning of reliability is clarified. It is argued that the reliability of an effective dose coefficient as a protection device can best be determined by comparing the nominal detriment adjusted cancer risk associated with the dose coefficient, with a best estimate of risk for the exposure pathway and exposed population group, taking into account uncertainties in biokinetic, dosimetric and risk parameters. The present work describes the application of parameter uncertainty analysis to quantify uncertainties resulting from internal exposures to ^{238}U , ^{226}Ra , ^{239}Pu , ^{241}Am , ^{137}Cs , ^{90}Sr , ^{131}I , ^{129}I , and ^3H by members of the UK public, confining consideration to uncertainties in biokinetic models and parameter values. The report does not consider uncertainties in risk directly, but derives uncertainties in the biokinetic models that are used to calculate the retention and excretion of radionuclides in the body, in order to calculate distributions of effective dose per unit intake. The central values and ranges of the distributions are used to inform the derivation of uncertainty factors (UF) for the different dose coefficients. A UF indicates a 95% probability of the risk coefficient being within a factor, UF, of the nominal risk associated with the appropriate ICRP dose coefficient, E_{50} , with respect to uncertainties in the biokinetic model and parameter values. The inferred UF values are around 2-3 for ingestion and 2-6 for inhalation for all age groups. It is instructive to consider these ranges alongside the likely levels of exposure that are expected from the radionuclides considered (micro-sievert range) and the dose limit for planned exposures for members of the public (1000 micro-sieverts).

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EXECUTIVE SUMMARY

Protection of workers and members of the public from ionising radiation is dependent on the assessment of radiation doses received. For radionuclides that have entered the body (“internal emitters”), doses must be determined indirectly using appropriate mathematical models that describe the distribution of radionuclides and the delivery of dose to body tissues over time. For this purpose, the International Commission on Radiological Protection (ICRP) publishes effective dose coefficients (effective dose per unit intake, Sv per Bq) for radionuclides that enter the body by inhalation and ingestion. These are used widely in radiation protection to ensure that exposures to internal emitters by radiation workers and members of the public are within prescribed dose limits, constraints or reference levels.

ICRP is clear on the intended use of equivalent and effective dose as protection quantities: they are to be applied as reference values without uncertainty. However, it is also recognised by ICRP and others that there are uncertainties in the process of estimating dose and risk that affect the derivation and application of these quantities. Although a number of publications describe the quantification of uncertainties on organ doses and the calculation of distributions of effective dose, which result from intakes of radionuclides, it is not clear how the calculation of these quantities relates to the intended use of dose coefficients as reference values. The issue of relevance for regulators and other stakeholders is not the magnitude of the “uncertainty” on dose estimates, but how “reliable” dose coefficients are for protection purposes, as a protection device. The issue of reliability and how it should be defined and assessed by quantifying uncertainties on dose and risk is the principal aim of the present report.

The ICRP system of protection, for both internal and external exposures to ionising radiation, is underpinned by nominal cancer risk coefficients. These are derived for hypothetical populations, and are age and gender averaged. The coefficients are not intended for risk assessment, but to support the use of effective dose as a protection device aimed at ensuring all exposures comply with dose limits and for comparison with constraints and reference levels. That the quantities ensure compliance can be tested by calculating the best estimate of risk for a given exposure pathway and then comparing this estimate with the nominal risk associated with the dose coefficient and expected level of intake. A dose coefficient is considered “reliable” if the magnitude of the difference between the best estimate of risk and nominal risk is small enough, with respect to the acceptable risk defined by the dose limit or constraint, that the dose coefficient is judged to be adequate as a protection device for the given exposure pathway. The concept of reliability therefore applies to the combination of dose coefficient and exposure pathway; consequently, the reliability of any given coefficient as a means of ensuring protection will be greatest in situations in which the models and parameter values used to calculate it are directly applicable to the circumstances of exposure. An overall assessment of reliability can be obtained by assessing the reliability of dose coefficients applied to some of the more significant exposure pathways.

The best estimate of risk for any exposure pathway is uncertain, and the quantification of this uncertainty is pre-requisite to making a judgement on reliability. It is shown that knowledge of the best estimate of risk and its uncertainty, for a given internal exposure pathway, is required to make an informed judgement on the reliability of a particular dose coefficient, as applied to a particular exposure pathway. Computational parameter uncertainty analysis can be applied to quantify this range as a probability interval using Monte Carlo methods. Where the effects of parameter uncertainty and variability can be precisely distinguished, the range can be calculated directly using a nested Monte Carlo method. In cases where this is not possible, conventional Monte Carlo can be used to calculate distributions of risk that reflect to varying extents the effect of parameter uncertainty and variability. These distributions can be used to infer the location of the risk coefficient with respect to the nominal risk. It is convenient to express uncertainty on this inferred range as an “uncertainty factor”, this indicates a 95% probability that the best estimate of risk for the population groups considered is within a factor, UF, of the nominal risk associated with the appropriate ICRP dose coefficient.

In this report, the consideration of uncertainties is limited to those inherent in the mathematical models that describe the distribution and retention of radionuclides in the body (biokinetic models). Uncertainties on model parameter values are derived and used to calculate distributions of effective dose per unit intake for 1 year old, 10 year old and adult members of the public, using parameter uncertainty analysis. Distributions are calculated for intakes that occur by ingestion and inhalation for ^{137}Cs , ^{90}Sr , ^{226}Ra , ^{238}U , ^{239}Pu , ^{241}Am , ^{129}I , ^{131}I and ^3H . The assumed exposure pathways are those that are considered likely to affect average members of the public (general environmental exposure), assuming that the exposure occurs in the United Kingdom. Ingestion is the dominant exposure pathway and exposure via this route is assumed to occur through the ingestion of radionuclides present in food and water, and to apply to radionuclides of natural or anthropogenic origin. Tritium is assumed to be consumed in the form of tritiated water (HTO), or tritium incorporated into commonly ingested organic material (organically bound tritium, OBT). For inhalation, exposure is assumed to occur to the following: methyl iodide vapour produced as a result of nuclear power generation or fuel reprocessing (^{129}I , ^{131}I); water vapour (^3H); uranium bearing aerosols consisting of various chemical forms of uranium produced in the nuclear fuel cycle or occurring naturally in the environment (^{238}U); residual aerosols produced from atmospheric nuclear tests and nuclear related accidents (^{137}Cs , ^{90}Sr , ^{238}U , ^{239}Pu , ^{241}Am); radium present in vitrified fly ash aerosols that result from the combustion of coal in coal fired power stations (^{226}Ra).

The calculated distributions are compared with dose coefficients that are deemed appropriate for assessing doses for the exposure pathways in question, in order to derive uncertainty factors. In this context, the derived UF values indicate a 95% probability that the best estimate of risk for the population groups considered is within a factor, UF, of the nominal risk associated with the appropriate ICRP dose coefficient, E_{50} , with respect to uncertainties in the biokinetic model. There are additional uncertainties in the dosimetry and risk models that will increase the UF values, although it is likely that uncertainties in the biokinetic model structure and parameter values, particularly those relating to material specific parameters, will dominate the uncertainties in dose and risk in most cases.

The derived UF values are given below and broadly indicate values of around 2-3 for ingestion, and 2-6 for inhalation. The UF values are the same for the three age groups considered: 1 year olds, 10 year olds and adults and are consistent with the geometric ranges of the distributions of dose being similar for each age group (absolute ranges of doses generally increased with decreasing age at exposure). The derived UF values are consistent with those inferred from other uncertainty studies. The UF values will be useful to regulators and others involved in applying radiation protection.

A general assessment of reliability of the protection quantities is beyond the scope of this report. However, the derived UF values for the radionuclides considered here are acceptable when considered alongside the likely levels of exposure that is expected from them (the sub micro-sievert to micro-sievert range) and the dose limit for planned exposures for members of the public: 1000 micro-sieverts (ICRP 2007); and considered in context with all radiological hazards to the general public: an estimated average annual dose of 2700 micro-sieverts per year in the UK, incurred mostly from radon and other natural sources of radiation, and medical exposures (Watson et al 2005).

The strategy described could be readily extended to include uncertainties in the estimation of cancer risk, and other parameters, for a more comprehensive assessment of uncertainty on risk and assessment of reliability of dose coefficients.

Radionuclide	Route of intake	Uncertainty Factor
¹³⁷ Cs	Ingestion	<3
¹³⁷ Cs	Inhalation	3
⁹⁰ Sr	Ingestion	3
⁹⁰ Sr	Inhalation	3
²³⁸ U	Ingestion	3
²³⁸ U	Inhalation	2
²²⁶ Ra	Ingestion	3
²²⁶ Ra	Inhalation	6
²³⁹ Pu	Ingestion	3
²³⁹ Pu	Inhalation	<3
²⁴¹ Am	Ingestion	3
²⁴¹ Am	Inhalation	<3
³ H (HTO)	Inhalation/ingestion	2
³ H (OBT)	Ingestion	2
¹³¹ I	Ingestion	2
¹³¹ I	Inhalation	2
¹²⁹ I	Ingestion	<4
¹²⁹ I	Inhalation	<4

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1 INTRODUCTION

Protection of workers and members of the public from ionising radiation is dependent on the assessment of radiation doses received. For radionuclides that have entered the body (“internal emitters”), doses must be determined indirectly using appropriate mathematical models that describe the distribution of radionuclides and delivery of dose to body tissues over time. These models and their parameter values are subject to uncertainty. Consideration of these uncertainties and how they affect the calculation of doses is important in the evaluation of the power of epidemiological studies and in assessments of risks from occupational and environmental exposures to radionuclides (Stayner et al 2007; CERRIE 2004; NCRP 2010). In addition, the quantification of uncertainties on doses is useful for understanding sources of uncertainty in the derivation of the protection quantity, effective dose.

The International Commission on Radiological Protection (ICRP) publishes effective dose coefficients (effective dose per unit intake, Sv per Bq) for radionuclides that enter the body by inhalation and ingestion (ICRP 1991, 2007). These are used widely in radiation protection to ensure that exposures to internal emitters by radiation workers and members of the public are consistent with prescribed dose limits, constraints or reference levels (ICRP 2007). A dose coefficient applies to a population group rather than to a specific individual. In this capacity, the coefficients are regarded as fixed values, and uncertainties are not considered in their application. Nevertheless, uncertainties are clearly present in the derivation of effective dose coefficients; contributors to these are:

1. The biokinetic model structures and their parameter values that are used to predict the dynamic distribution of radioactivity within the body.
2. The geometric relationship of source and target tissues, their dimensions and masses; these influence the amount of energy deposited in tissues.
3. The relative effectiveness of different radiation types in causing cancer and differences between tissues in their sensitivity to radiation-induced health detriment.
4. The extrapolation of risk estimates to different populations and the assumption of linearity in the dose response relationship (the Linear No Threshold model)

Because effective dose coefficients apply to sex-averaged reference persons calculated using chosen radiation and tissue weighting factors they are not rigorous scientific quantities that can be regarded as best estimates subject to uncertainties (ICRP 2007, Harrison and Day 2008). Instead they should be regarded as a radiation protection tool for use in the control of sources of exposure. However, an important conclusion of a UK government committee, which reviewed risk to the public from internal emitters, was that this position can only be justified by the consideration and quantification of the uncertainties inherent in the derivation of dose coefficients (CERRIE 2004). Thus, although ICRP publishes dose coefficients as fixed quantities, their reliability as a means of ensuring protection will be greatest in situations in which the models and

parameter values used in the calculations are based on good data that are directly applicable to the circumstances of exposure.

Several different approaches to quantification of uncertainties on doses have been applied, one of the most useful being parameter uncertainty analysis. Here probability distributions, which represent the state of knowledge of the model and its parameters, are propagated through the calculation of dose using a Monte Carlo method (NCRP 2010); such a process produces a distribution of doses. The method has been applied to calculate distributions of effective dose per unit intake for a number of radionuclides and routes of intake. However, although useful for identifying important sources of uncertainty, it is not clear how distributions of effective doses should be interpreted with respect to the intended use of effective dose in radiation protection. The issue that is of importance to regulators and other stakeholders is not the magnitude of the “uncertainty” on dose estimates, but how reliable dose coefficients are as a protection device.

It should be pointed out that reliability relates to a particular exposure scenario and dose coefficient. For example, the dose coefficient for inhalation of Type M (medium lung solubility) forms of plutonium will be more reliable when used to assess doses from inhalation of compounds such as plutonium nitrate, for which the lung solubility parameter values have been determined from in vivo studies and shown to be consistent with Type M behaviour (Davesne et al 2010, Puncher et al 2011), than when applied to assess doses from a chemical form whose identity is unknown, or has not been studied in vivo but is thought to behave like a Type M material.

1.1 Aims and scope of the report

The objectives of this report are to:

- Review the ICRP system of protection and clarify the concept of “reliability” as it relates to dose coefficients and their application in prospective internal radiation protection.
- Identify how reliability can be determined and how the quantification of uncertainties on dose and risk using parameter uncertainty analysis can be used to inform this process.
- Perform uncertainty analysis to calculate distributions of doses (effective dose per unit intake) from ingestion and inhalation for a range of important radionuclides: ^{137}Cs , ^{90}Sr , ^{226}Ra , ^{238}U , ^{239}Pu , ^{241}Am , ^{129}I , ^{131}I , ^3H . The analyses are performed for 1 year old, 10 year old and adult members of the public as defined by ICRP (ICRP 1995).
- Use the calculated distributions of dose to derive “uncertainty factors” for each dose coefficient that can be used to assess reliability. The uncertainty factors are compared with those inferred from the results of previous studies.

2 THE RELIABILITY OF DOSE COEFFICIENTS

2.1 The ICRP system of protection

Effective dose is central to the ICRP radiation protection system. ICRP publishes dose coefficients (Sv Bq^{-1}) for inhaled or ingested radionuclides, giving both equivalent and effective dose (ICRP 1991, 2007). The steps in the calculation are shown in Figure 2.1 and can be summarised as follows:

- Biokinetic models are mathematical representations of the dynamic distribution of radionuclides in the body, and are used to calculate the number of disintegrations that occur in source regions of the body (organs and tissues).
- Dosimetric models define the geometric relationship between source and target organs. They are used to calculate the absorbed dose (Gy) received by every target organ resulting from radioactive disintegrations that have occurred in source organs.
- Radiation weighting factors, w_R , are used to take account of the relative effectiveness of different radiation types in causing cancer and hereditary effects at low doses and dose rates, converting absorbed dose to equivalent dose, H_T (Sv).
- Tissue weighting factors, w_T , are used to further weight equivalent doses, H_T , to individual organs and tissues according to their relative contribution to health detriment, to produce a weighted equivalent dose for each organ or tissue.
- Effective dose, Sv per Bq intake, is obtained by summing the weighted equivalent organ and tissue doses.

A detailed explanation of the calculation of effective dose can be found in the most recent ICRP recommendations (ICRP 2007); Harrison and Day (2008) provide a summary and a commentary on the ICRP quantities and their intended application. The use of effective dose allows the summation of the dose from different radionuclides and from external sources for comparison with dose limits and constraints set on the basis of risks relating to whole-body radiation exposure. Equivalent and effective dose coefficients published by ICRP are integrated over a 50 year period for adults and to age 70 years for children and the resulting values are referred to as committed doses.

2.1.1 ICRP nominal risk coefficients

ICRP publishes sex and age averaged, nominal cancer risk coefficients (the detriment adjusted radiation induced excess absolute cancer incidence incurred per sievert of exposure) for members of the public and workers. These are derived from estimates of cancer risk obtained largely from follow-up studies of the survivors of the atomic bombings at Hiroshima and Nagasaki in which people were exposed primarily to acute low linear energy transfer (LET) external radiation. The risk coefficients were obtained as follows:

1. Lifetime cancer incidence risk estimates were derived for 14 organs and tissues, and gender averaged.
2. The risk estimates for solid cancers were adjusted downward using an empirical dose and dose-rate effectiveness factor (DDREF).
3. The risk estimates were transferred across populations to obtain nominal risk coefficients that are averaged over seven western and Asian populations.
4. The risk coefficients were further adjusted for cancer lethality, loss of quality of life (which accounts for the differential morbidity and suffering associated with non fatal cancers) and years of life lost.

ICRP has derived a nominal cancer risk coefficient of 5.5% per sievert for the “whole population” and 4.1% per sievert for the “adult population” (ICRP 2007). These populations do not model any specific population group; at best it can be said they are representative of the world population. In addition, the tissue weighting factors, which broadly represent the apportionment of the nominal risk among radiosensitive body organs and tissues, are rounded, sex and age averaged quantities. For these reasons it is inappropriate to use effective dose as a surrogate for risk when assessing exposures to radionuclides for particular individuals or population groups (ICRP 2007, Harrison and Day 2008). Instead, the nominal coefficients are intended to support the use of effective dose as a protection device aimed at ensuring all exposures comply with dose limits and constraints.

In situations where an estimate of the risk incurred from exposure to ionising radiation is required, the use of effective dose is inappropriate and ICRP (2007) recommend that a suitable risk assessment should be performed for the population group or individual in question. Such an assessment would utilise risk coefficients that are representative of the exposed population or person in terms of age at exposure, ethnic origin and gender.

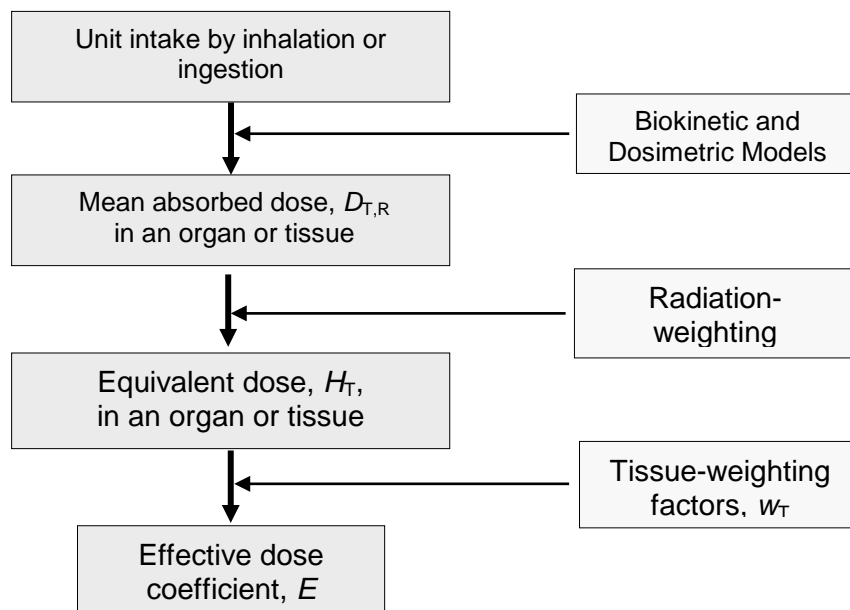


Figure 2.1. The steps in the derivation of effective dose coefficients.

2.2 Assessing the reliability of dose coefficients

A risk coefficient for a particular radionuclide and route of intake predicts the average (expected) lifetime risk of developing a cancer, per unit intake of the radionuclide, for the population in question. Thus, if every member of the population experienced a unit intake of the radionuclide, and the risk coefficient for the population in question is known accurately, then the average number of individuals incurring cancer will be the product of the number of individuals in the population and the risk coefficient.

The population in this context could be a large heterogeneous group of individuals, in terms of age and gender, or a defined sub-group. An example of the latter could be the hypothetical “representative person” identified in a particular environmental exposure pathway (ICRP 2006a). The general steps in the derivation of the best estimate of the risk coefficient for any population group is broadly similar to the calculation of effective dose (Figure 2.2); the specific steps are described further in Appendix A.

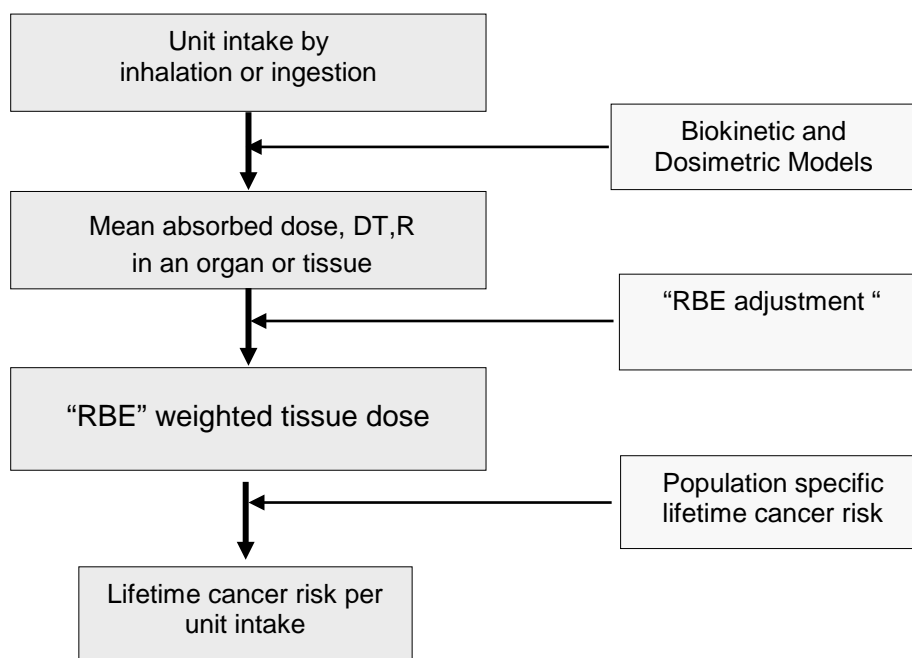


Figure 2.2 The steps in the derivation of population specific risk coefficients.

It might be argued therefore that the “ideal” protection quantity for any particular radionuclide and exposure pathway, and exposed population group, should be the risk coefficient that gives the best estimate of detriment adjusted excess absolute cancer incidence per unit intake, to the population in question. However, the derivation of such a quantity for every exposure scenario is not feasible for a workable system of radiation protection. Instead, quantities are required that provide nominal estimates of risk that are adequate for ensuring exposures are within acceptable levels. Because anticipated exposures are generally much lower than dose limits, it is expected that a reasonable difference between the value of the nominal and best estimate of risk is acceptable for most radionuclides and routes of exposure. Nevertheless, quantifying this difference provides a means of testing this assumption for any particular dose coefficient and

exposure pathway, and so assessing the “reliability” of effective dose coefficients as a protection device.

The process is illustrated in Figure 2.3. The risk (the best estimate of the detriment adjusted cancer incidence for the population) for the exposure pathway and population in question is the product of the expected level of intake for the pathway in question, I , and the best estimate of the population specific risk coefficient, R ; the corresponding nominal risk is the product of the intake and the nominal risk associated with the dose coefficient, R_{E50} . Reliability is ascertained by comparing the values of the actual risk ($I \times R$) and nominal risk ($I \times R_{E50}$), and the risk defined by the prescribed dose limit or constraint for members of the public, (R_{DL}).

A dose coefficient can be considered reliable as a protection device if the following conditions are met:

1. The best estimate of risk ($I \times R$) is less than the risk associated with the dose limit (R_{DL}).

- 2.

If the best estimate of risk ($I \times R$) is greater than the nominal risk ($I \times R_{E50}$), then the magnitude of the difference between the best estimate of risk and nominal risk, D_E , and the difference between the best estimate of risk and risk associated with the dose limit, D_R , is judged to be small enough that it can be ignored for radiation protection purposes.

Or

If the nominal risk ($I \times R_{E50}$) is greater than the best estimate of risk ($I \times R$), then the magnitude of the difference between the best estimate and nominal risk, D_E , is not so large that ensuring exposures comply with dose limits and constraints is unduly cautious.

3. The uncertainty on the risk coefficient is small enough to permit conditions 1&2 to be evaluated.

Point 3 highlights the fact that the reliability of a dose coefficient as a protection device in a particular exposure scenario depends on how precisely the risk coefficient can be determined; this is often dependent on how much information is available concerning the exposure. For example, if an intake by inhalation is suspected, but the chemical form of the material is unknown or cannot be inferred, then the uncertainty on the risk coefficient will be significant because the material specific parameters that affect the calculation of dose, and hence risk, can assume a wide range of plausible values.

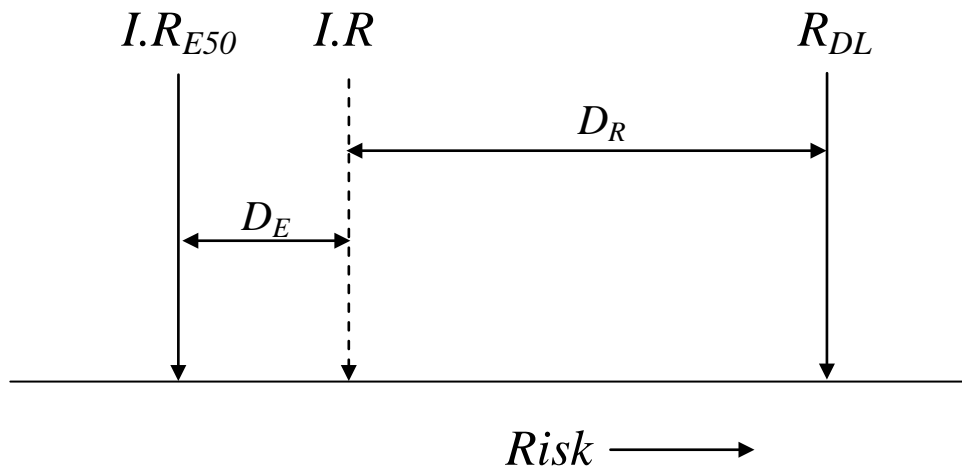


Figure 2.3. Assessing the reliability of dose coefficients. The axis denotes detriment adjusted cancer risk to a hypothetical population. R is the risk coefficient (risk per unit intake) for some radionuclide of interest; R_{E50} is the nominal risk associated with the dose coefficient for the same radionuclide; R_{DL} is the risk associated with the prescribed dose limit. The product, $I.R$, is the magnitude of the risk incurred to the population from the expected (average) intake, I , of the radionuclide for some exposure pathway of interest; $I.R_{E50}$ is the nominal risk. The reliability of the dose coefficient can be determined by comparing the actual ($I.R$) and nominal risks ($I.R_{E50}$) with the risk set by the dose limit (R_{DL}) (see main text).

2.3 Uncertainty analysis

The role of uncertainty analysis in assessing reliability is to quantify the uncertainty on the range of values of the risk coefficient. Because the risk coefficient represents a population average, it is uncertainty on the best estimate of the mean value that is required. The uncertainty on the mean value should not be confused with stochastic variation (the frequency distribution) of the risk coefficient in the population of interest. The uncertainty on the mean provides a means of inferring the range of values of D_E to assess the reliability criteria noted above.

2.3.1 Uncertainty and Variability

It is important to distinguish “uncertainty” from “variability” when calculating a mean value. Uncertainty refers to the lack of knowledge regarding a specific parameter value; the value itself is fixed but is not known with precision. This type of uncertainty is expressed as a probability in Bayesian inference or a confidence interval in classical statistics (Spiegelhalter et al 2004). Relevant examples of “uncertain” parameters include the values derived for rate constants in the biokinetic models that predict the time dependent distribution of radionuclides in the human body; these are usually regarded as central or mean values in the human population of interest. Another example would be uncertainty on the chemical form of an inhaled radionuclide, or the precise values of the lung solubility parameters of the material where the exposure is to a known chemical form. On the other hand, variability refers to the natural range and frequency that a parameter value can assume in individuals within the population of interest. Relevant examples of “variability” include inter-individual variation in the fraction of an ingested radionuclide taken up to blood from the alimentary tract, or natural variation in the mass of the liver between individuals in a population. An important point is that if the distribution of a variable parameter is known with absolute

precision then there would be nothing “uncertain” about it. In other words, it is a frequency distribution and not a probability distribution. In reality of course, estimates of the values of variable parameters, where measurement or indirect inference is possible, will be uncertain so that the obtained distribution will reflect a combination of measurement uncertainty and variability in the parameter value. Variability and uncertainty are sometimes referred to as aleatory and epistemic uncertainty, respectively. A discussion of the two types of “uncertainty” and their implications for internal dose assessment is provided by NCRP (2010). A detailed discussion of uncertainties is provided by ISO (1993).

Parameter uncertainty will result in a systematic increase or decrease in the population mean risk coefficient. The extent of this effect will depend on how sensitive the calculation is to the parameter in question. To properly account for the effect of uncertainty in a parameter uncertainty analysis, the probability distribution that represents the state of knowledge of the uncertain parameter must be propagated directly into the distribution that represents uncertainty on the population risk coefficient using the Monte Carlo method. However, if parameter variability is misinterpreted as uncertainty, then the distribution of coefficients resulting from an uncertainty analysis that propagates this frequency distribution (misclassified as an uncertainty distribution) through to the distribution of coefficients, will overstate the uncertainty on the population mean. This is because parameter variability does not directly affect the variance of the mean, but may affect its location if the model is non-linear with respect to the variable parameter or the frequency distribution representing variability in the parameter is asymmetric. A simple example of the effect of the latter is provided by the example of uranium absorption from the alimentary tract. If the risk model reference value for the f_1 parameter, which predicts the fraction of uranium taken up to blood from the alimentary tract, is the median value of a log-normal distribution, with geometric standard deviation (GSD) of 2, which represents variability in uranium absorption in the population of interest, then the population mean risk coefficient would be shifted by a factor $\sim \exp(\ln(2)^2/2) = 1.3$, with respect to the model prediction. The effect becomes more pronounced as the GSD increases.

2.3.2 Uncertainty factors

Puncher and Harrison (2012a) describe a nested Monte Carlo method to estimate the uncertainty on the risk coefficient; this produces a distribution of mean values that can be used to directly determine ranges for the value of D_E that are required to assess the reliability of dose coefficients. The method mathematically distinguishes the effects of parameter uncertainty and variability. In practice, however, it is often difficult to distinguish uncertainty from variability accurately enough to justify use of the method when deriving distributions for model parameter values, particularly when data are sparse.

An alternative to using the nested Monte Carlo approach is to use the standard Monte Carlo method to propagate derived distributions of model parameters directly into the distribution of dose. Under these circumstances, the distributions do not reflect accurately uncertainty on the mean because they also include a variance term introduced by variability. In other words, the distributions may be “wider” or even biased,

because this variance term is not integrated out when the uncertainty analysis is performed. Although the distributions that are calculated using this method do not provide a direct measurement of the uncertainty on the value of D_E , the relative contribution to the total variance that results from uncertainty and variability in parameter values can be assessed subjectively and the distributions used to *infer* a plausible range of D_E . This subjective range can be expressed as an uncertainty factor, UF, which provides a geometric range for the likely values of D_E . The UF indicates that the risk coefficient has a 95% probability of being within a factor, UF, of the nominal risk associated with the dose coefficient. This is consistent with the use of subjective “uncertainty factors” described elsewhere (Harrison et al 2001, Leggett et al 2001).

3 UNCERTAINTY ANALYSIS

As discussed in section 2, the role of uncertainty analysis should be to quantify the range of the best estimate of risk coefficients in order to make comparisons with the nominal values feasible. It is clearly not possible, however, to use this approach to assess the reliability of dose coefficients in every conceivable exposure scenario. Instead, an overall assessment of reliability can be obtained by limiting the described approach to dose coefficients that are used to assess doses from the most important exposure pathways: those that give the highest doses to the general population or are of concern to the public because the radionuclides are of anthropogenic origin. With this in mind, the present report describes the application of parameter uncertainty analysis to quantify uncertainties resulting from intakes of radioisotopes of uranium, plutonium, americium, strontium, caesium, iodine and tritium, occurring by ingestion and inhalation through the most likely exposure pathways. With regard to the latter, the work considers general routes of exposure that affect members of the public in the United Kingdom.

The consideration of uncertainties is restricted to those inherent in the structure of the biokinetic models and parameter values, so that the output of the Monte Carlo calculations are distributions of effective dose per unit intake calculated using the ICRP Publication 60 formalism (ICRP 1991). These distributions, which reflect, to varying extents, uncertainty and variability in parameter values are used to infer the uncertainty on the mean effective dose per unit intake to derive UF values for the appropriate dose coefficients for ingestion and inhalation that are currently applied to protect members of the public. In this context, the derived UF values indicates a 95% probability that the best estimate of risk for the considered population groups is within a factor, UF, of the nominal risk associated with the appropriate ICRP dose coefficient, E_{50} , with respect to uncertainties in the biokinetic model. Note that there are additional uncertainties in the dosimetry and risk models that will increase the UF values, although it is likely that uncertainties in the biokinetic model structure and parameter values, particularly those relating to material specific parameters, will dominate the uncertainties in dose and risk.

3.1 Radionuclides and routes of exposure

3.1.1 Uranium

Exposures to uranium in the UK occur mainly to naturally occurring ^{238}U and ^{234}U , around 85% of the dose received occurs via the ingestion pathway (Watson et al 2005). Exposures to uranium discharged to the environment can also occur near nuclear establishments involved in fuel fabrication and reprocessing (EA 2010). Exposure via ingestion occurs via low level chronic exposure to soluble material ingested in food and water. For inhalation, exposures to uranium aerosols are most likely to occur via inhalation of aerosols containing various chemical forms, principally oxides, including uranium dioxide (UO_2), uranium trioxide (UO_3) and uranium octoxide (U_3O_8).

3.1.2 Radium

Exposures to radium in the UK occur mainly via ingestion of naturally occurring ^{226}Ra in food and water, formed from the decay of ^{238}U (Watson et al 2005). A secondary route of exposure occurs through inhalation of ^{226}Ra present in aerosols bearing fly ash produced from the combustion of coal in coal fired power stations (Smith et al 2001).

3.1.3 Caesium and strontium

Exposures to caesium and strontium occur via ingestion of food and water containing trace amount of ^{137}Cs and ^{90}Sr released into the environment from nuclear power generation and fuel reprocessing. Exposures via inhalation, a secondary route of exposure, occurs to residual fission products containing ^{137}Cs and ^{90}Sr , formed as a result of nuclear weapons tests conducted prior to 1964, and to aerosols produced from nuclear accidents, particularly the Chernobyl incident (Watson et al 2005). Inhaled material is in aerosol form and likely to be fairly soluble.

3.1.4 Plutonium and americium

Exposures to plutonium and americium occur via ingestion of food and water containing trace amounts of $^{239/240}\text{Pu}$ and ^{241}Am released from nuclear fuel reprocessing. Exposures via inhalation are a secondary route of exposure and likely result from exposure to aerosols bearing plutonium and americium in insoluble oxide form. The aerosols are primarily the residue from atmospheric nuclear tests conducted up to 1964.

3.1.5 Iodine

Exposures to both ^{131}I and ^{129}I can occur in the UK; the former is released into the atmosphere as a result of nuclear power generation; the latter from fuel reprocessing. Ingestion occurs via the presence of these radionuclides in food and water. Inhaled material is likely to be gaseous, with methyl iodide gas being the primary form (Collins et al 2004).

3.1.6 Tritium

Exposures to tritium occur via the ingestion (the dominant pathway) and inhalation of tritiated water (HTO); and also to organically bound tritium present in food (tritium associated with carbohydrate, fat and protein. Tritium is produced as a by-product of the nuclear fuel cycle.

3.2 Biokinetic models and model uncertainty

Mathematical models are used to determine the distribution of inhaled and ingested radionuclides to the different tissues of the body, and their clearance from these tissues and ultimate removal from the body in excreta (ICRP 1993, 1995a). These models are often referred to as “biokinetic models”, and can be subdivided into:

1. The models that govern deposition and clearance to blood from the respiratory tract (intakes by inhalation) or alimentary tract (intakes by ingestion); these are sometimes referred to as “intake tract” models. The structure and the values of physiological parameters (parameters that are assumed to be independent of the chemical form of the ingested or inhaled material) are assumed to be radionuclide independent, and are thus applied to calculate doses from intakes of all radionuclide species. Parameters that govern absorption from the lungs or alimentary tract are assumed to be material or chemical specific and therefore are assumed to have the same values for different individuals.
2. The models that govern the clearance of radionuclides from blood (after absorption from the lungs or alimentary tract) to systemic tissues, their retention in tissues and excretion from the body; these are termed “systemic models” and are radionuclide and sometimes chemical specific.

A detailed description of the biokinetic models considered in this report is provided in Appendix B and summarised below.

3.2.1 The alimentary tract model

The analyses used an earlier alimentary tract model published in ICRP Publication 30 (ICRP 1979) that was used to calculate the ICRP 68 and 72 dose coefficients for workers and members of the public (ICRP 1994a, 1995a). A revised model was published in 2007 (ICRP 2007). However, with respect to radionuclide biokinetics, the revised model reduces to the older form for most radionuclides and the ones considered in this report.

The most important parameter in either model is the fraction of ingested activity that is absorbed to blood from the alimentary tract: it ultimately determines the amount (and hence dose) of ingested radionuclides that are deposited in tissues of the body. In ICRP Publication 30 this parameter is termed the f_1 value and is assumed to be chemical and age dependent. The same assumption is made here.

3.2.1.1 Age dependent modifications to the alimentary tract model

In the ICRP Publication 30 model, the transit rates through the alimentary tract are assumed to be the same for all age groups. ICRP notes that this assumption is likely to overestimate doses to regions of the alimentary tract for infants and children (ICRP 1979). The same values are assumed for all age groups in this analysis.

3.2.2 The human respiratory tract model (HRTM)

All models that attempt to determine doses to body tissues (be it chemical or radiological) that result from inhalation of pollutants or radionuclides, incorporate some form of representation of the lung. In toxicokinetic modelling, this is often simply a compartment representing deposition and clearance from the alveolar region (Reddy et al 2006). In internal radiation protection, however, several models have been developed of varying complexity, which model the regional deposition and clearance of inhaled radionuclides from the lungs (ICRP 1979, 1994b; NCRP 1997). Of these, the ICRP Publication 66 human respiratory tract model (ICRP 1994b) is probably the most widely applied to model exposures to radionuclides occurring by inhalation. The model is used to estimate the deposition, clearance and regional dosimetry of inhaled radionuclides in aerosol and vapour form (ICRP 1994b). It was derived with modularity in mind: it can link directly to available systemic and alimentary tract models to determine doses to all tissues that result from exposure via inhalation. An alternative model has also been published by the National Council on Radiation Protection and Measurement (NCRP 1997). Use of this model was not considered in this analysis because:

A. The ICRP Publication 66 model is considered to represent a sufficient level of physiological and physical realism for internal dosimetry purposes: for example, it is used to reconstruct lung doses for plutonium workers in epidemiology studies (Khokhryakov et al 2005, Puncher et al 2011) in addition to being used in applied radiation protection, and has also been used for toxicological risk assessment resulting from intakes of inorganic pollutants (ATSDR 2011);

B. The model has been revised to account for more recent data describing deposition and clearance of inhaled radionuclides from the airways of the lung (Bailey et al 2007; Gregoratto et al 2010; Smith et al 2007, 2011). These revisions were included in the present study and are described in Appendix B.

3.2.2.1 Age dependent modifications to the Human Respiratory Tract Model

Due to an absence of age specific data, the rate constants governing the clearance of material from the lungs by particle transport and absorption are assumed to be age and sex independent in the HRTM. However, the model accounts for age and sex specific variation in the deposition of inhaled aerosols, which are accounted for by the differences in the dimensions of airway walls and the breathing habits of different age groups and genders. The inclusion of age and sex specific mathematical phantoms and target organ masses account for dosimetry differences for penetrating emissions (ICRP 1994b).

3.2.3 Systemic models

Various models have been developed to calculate the distribution of radionuclides to body tissues, and their subsequent removal, after uptake to blood. The majority of these were developed for or by ICRP for the purpose of calculating dose coefficients (ICRP 1994a, 1995a). Because the ICRP models generally provide a sufficient level of realism and were developed to calculate doses for different age groups that consider, for example, age dependent differences in biokinetics such as bone metabolism, the uncertainty analyses were conducted using the ICRP model. However, in cases where an alternative model structure was available, the likely effect of the model on the derived uncertainty factor was considered and, for some radionuclides, additional parameter uncertainty analyses were performed using the alternative model structure. The systemic models used in the analysis are summarised below; further discussion is provided in Appendix B.

3.2.3.1 Strontium and radium

The ICRP systemic models for strontium and radium, which are based on a generic structure derived for the alkaline-earth metals, and physiological analogues lead and uranium, were used in the analyses (ICRP 1993; Leggett 1992a). An alternative model for radium has been published by Polig et al (2004), which is arguably less realistic and does not consider age dependent dosimetry. In any event, it gives similar values of effective dose per unit ingestion (as calculated by the authors) to the ICRP model.

3.2.3.2 Uranium

The ICRP Publication 69 model for uranium was used, which is based on the generic structure for alkaline-earth metals published by Leggett (1992a).

3.2.3.3 Plutonium and Americium

The ICRP Publication 67 models for plutonium and americium were used (ICRP 1993, Leggett 1992b). An updated model for plutonium, based on the ICRP Publication 67 model, has been published by Leggett et al (2005) for adults. Uncertainties on doses from inhalation for adults, calculated using the older systemic model, are compared with additional calculations performed using the revised model; the results from ingestion are compared with a previous analysis conducted using the revised model by Puncher and Harrison (2012b).

3.2.3.4 Iodine

The analyses were performed for all age groups using the ICRP Publication 56 model for iodine (ICRP 1989). These are compared with additional calculations for adults using an updated model published by Leggett (2010).

3.2.3.5 *Caesium*

The analyses were performed for all age groups using the ICRP Publication 56 model for caesium (ICRP 1989). These are compared with additional calculations for adults using a more physiologically realistic model, based on one published by Leggett et al (2003).

3.2.3.6 *Tritiated water (HTO) and organically bound tritium (OBT)*

The analyses were performed for all age groups using the ICRP Publication 56 models for HTO and OBT (ICRP 1989). Physiologically more realistic alternatives are discussed in section 3.4.3.6, and the results of the analyses are compared with similar analyses published by others using the alternative models.

3.3 Derivation of uncertainties

A complete description of the derivation of distributions of model parameter values is provided in Appendix C. This section summarises the important parameters, the rationale behind the choice of distributions, and whether the derived distributions are considered to reflect mainly “uncertainty” (directly affects the uncertainty on dose) or “variability” (affects the location of the distribution of uncertainty on dose with respect to the corresponding ICRP value).

3.3.1 The alimentary tract model

3.3.1.1 *The fraction of activity absorbed to blood from the alimentary tract, the f_1 value (uncertainty).*

With the exception of tritiated water (HTO), whose rate of absorption to blood was assumed to be instantaneous, a distribution for the f_1 value was derived for each radionuclide, considering age dependent differences in absorption from the alimentary tract. Because of the near linear relationship between the value of this parameter and dose, the distributions were derived to represent uncertainty on the population mean value.

3.3.2 The human respiratory tract model (HRTM)

The magnitude of the effective dose per unit intake resulting from inhalation of radionuclides is dependent on:

1. The amount of activity deposited in the lung (in aerosol or vapour form), and the regional lung deposition pattern
2. The rate of clearance of material from the lungs to blood (absorption) or to the alimentary tract and lymph nodes (particle transport clearance).

3.3.2.1 Deposition

Aerosols (variability)

With the exception of iodine, which is assumed to be inhaled as vapour, all radionuclides were assumed to be inhaled in aerosol form. The amount and pattern of aerosol deposition in the lungs is dependent on the particle size distribution of the aerosol: assumed to be a lognormal distribution defined by a median value (the activity median aerodynamic diameter, AMAD) and dispersion parameter (a geometric standard deviation, GSD) - and the breathing pattern assumed at the time of intake (ICRP 1994b). Previous studies have shown that variation in lung dose and effective dose per unit intake is less sensitive to variation in the deposition parameters compared with the parameters that determine lung clearance; the exception being the fraction of inhaled activity breathed through the nose, which can influence the amount of inhaled activity reaching the lungs (Puncher et al 2008, Puncher et al 2011). The breathing parameters, including the fraction of material breathed through the nose, largely reflect inter-subject variation in the breathing pattern among individuals. In this study, the breathing pattern was assumed to vary about the values assumed for the age dependent time spent in different daily activities: sleeping, sitting, light and heavy exercise.

The size distributions of environmental aerosols are variable, depending on the process of formation. Given the secondary effect of the size distribution parameters on dose, and in the absence of more specific information, the same distributions were assumed for the location and dispersion parameters for all radionuclide exposures occurring in aerosol form. The broad distribution encompasses the variation observed among different types of environmental aerosols collated by Dorrian (1997).

Vapour (uncertainty)

Iodine was assumed to be inhaled in vapour form, primarily as methyl iodide gas. The amount of methyl iodide vapour that is deposited in the lung has been determined from volunteer studies (ICRP 1995a). Because there is a near linear relationship between this fraction and effective dose per unit intake (it is assumed to be instantaneously absorbed to blood from the lungs), the distribution derived for this parameter reflected uncertainty in the mean value among the population.

3.3.2.2 Clearance

Particle transport clearance (variability)

Aerosol particles and vapours deposited in the airways are cleared primarily to the back of the throat and swallowed. In the upper thoracic airways (the tracheo-bronchial region), clearance is mediated by the mucociliary escalator; clearance from the alveolar-interstitial (AI) region to the tracheo-bronchial region (and subsequent removal by mucociliary clearance) and thoracic lymph nodes is mediated by macrophage activity. The particle transport clearance pathway is assumed to be independent of the chemical or physical form of the inhaled material. In this analysis, the distributions derived for the rates of particle transport clearance were assumed to represent primarily inter-subject variation in the rate of this clearance pathway between individuals, and hence are a source of variability.

Absorption (uncertainty)

The solubilisation of inhaled particles in the airways and subsequent uptake to blood is a competing clearance process to particle transport. Soluble materials tend to give higher doses to systemic tissues, insoluble materials higher doses to lung tissues. The rate of absorption is assumed to depend on the chemical or physical form of the inhaled material: for example the radionuclide may be in an insoluble chemical form or buried in an insoluble matrix. If the exposure pathway is dominated by a single type of material (e.g. environmental aerosols containing plutonium are likely to be in oxide form), then uncertainty in the rate of absorption of the material systematically affects the dose to all individuals in the population. With the exception of uranium, this report assumes that exposures to radionuclide bearing aerosols and vapours occur via exposure to the same chemical (e.g. plutonium oxide) or physical form (e.g. radium embedded in vitrified fly ash); this was assumed to be a source of uncertainty. However, for uranium, exposure was considered more likely to occur to a mixture of chemical forms; this is a form of variability. For all materials, the distribution values of absorption parameters were based on a literature review of the clearance of relevant materials from the lungs.

3.3.3 Systemic models

To derive distributions for rate constants in the systemic models, sensitivity analyses were performed to identify rates having the greatest impact on dose per unit intake. This process is described in Appendix C.

Lognormal distributions were chosen to represent variation in parameter values. These were chosen because:

- A. Many rate constants that influence effective dose have values that are close to, but are constrained to be greater than, zero.
- B. The form of the distribution has been shown to be relatively unimportant for calculating distributions of effective dose per unit intake if the central value and inter-percentile ranges in the distributions are similar (Puncher and Harrison 2012b).

In most cases the median of the lognormal distribution was assumed to be the reference rate in the model. This was justified on the basis that the derived model parameters reproduce the median values of the human bioassay data the model was fitted to and are often referred to, albeit loosely, as “central” values.

For all radionuclides except tritium and iodine, distributions were assigned to parameters that reproduced the range of organ retention or excretion data (bioassay data) observed in the human population. In this approach, an initial distribution was chosen for each parameter; these were then adjusted in an iterative fashion until the 95% range of predicted retention of activity in the organs of interest or excreta reasonably reproduced the range of published human data. In the case where the rate did not appreciably affect bioassay data, for instance, rates within skeleton, wide distributions were chosen and additional analyses that did not include varying these parameters were performed to assess their effect on the distribution of dose. The distributions were assumed primarily to represent a source of variability. In the absence

of specific data, the same dispersion about the median values was assumed for each age group.

For iodine and tritium, the derived distributions represented uncertainty on the mean values of systemic parameters; this was justified on the basis that the systemic rates that determine effective dose are well characterised from human data (the individual rates are directly measurable). For iodine, effective dose is dominated by the dose to the thyroid which is determined by the rate of uptake of iodine by the thyroid from blood (^{131}I and ^{129}I) and the rate of removal from the thyroid (^{129}I). For tritium, effective dose is determined by the rate of removal of activity from two biological compartments: 1. Body water (HTO) - short term retention; 2. Tritium associated with organic material (OBT) – long term retention. In the case of iodine, the distribution representing uncertainty on the rate of uptake of iodine by the thyroid was centred on a value higher than the ICRP value. This was justified on the basis that the value of this rate is inversely related to the amount of dietary iodine and it has been shown that the UK population has a mild iodine deficiency (Vanderpump et al 2010).

3.4 Uncertainty analysis

3.4.1 Monte Carlo Methods

Monte Carlo calculations were performed using a software tool developed to analyse uncertainties on doses (Puncher and Birchall 2007). This code samples parameter values from defined distributions and is interfaced with appropriate dosimetry codes (Birchall et al 2007; Fell 2007).

The Latin Hyper-cube sampling algorithm (McKay et al 1979) was used to sample parameter values. A Latin Hyper-cube matrix of 500 variates was constructed for model parameters using the derived distributions; up to three separate simulations were performed to monitor convergence of the calculated distributions of dose. In each simulation, the following steps were performed for every vector of sampled parameters following an acute ingestion or inhalation of 1 Bq of ^{137}Cs , ^{90}Sr , ^{238}U , ^{226}Ra , ^{239}Pu , ^{241}Am , ^3H (as HTO or OBT), ^{131}I or ^{129}I :

- The parameter values were set in the dosimetry code.
- Bioassay predictions were calculated at logarithmically spaced intervals up to 10,000 days after an acute intake, typically at times $T = 10, 20, 50, 100, 500, 1000, 2000, 5000$ and 10,000 days after the intake; predictions were calculated for all systemic organs including blood, kidneys, massive soft tissues, skeleton, liver, and 24-hour urinary and faecal excretion (Bq d^{-1}).
- The committed effective dose, $E(50)$ was calculated using ICRP Publication 60 radiation and tissue weighting factors (ICRP 1991).

The above steps were repeated for each of the 500 vectors of parameters in the Latin Hypercube matrix.

3.4.1.1 *Sensitivity analysis for doses from inhalation and ingestion*

With the exception of caesium (ingestion), tritium and iodine, further Monte Carlo calculations were performed for adults to determine which parameters, or combinations of parameters, were primarily responsible for the observed variation in doses from ingestion or inhalation. Further calculations were not performed for ingestion of caesium because the range of doses was deemed small enough that it was considered unnecessary to assess the contribution of uncertainty and variability to the calculated dispersion in dose; for tritium and iodine the distributions of parameter values were assumed to reflect only uncertainty.

Sensitivity of dose to the f_1 parameter (ingestion)

Additional Monte Carlo calculations were undertaken to assess the relative contribution that variation in the systemic parameters, and the f_1 parameter, made to the overall uncertainty on effective dose resulting from ingestion. To do this, the Monte Carlo procedure for adults was repeated with the following modifications:

1. The systemic parameters were varied but the f_1 value was fixed at the median value (lognormal and uniform distributions) or modal value (triangular distributions).
2. The systemic parameters were fixed at the median values of the parameter distributions, but the f_1 value was varied.

Sensitivity of dose to the lung absorption parameters (inhalation)

Further Monte Carlo calculations were undertaken to determine the contribution that uncertainties on absorption parameters made to the overall uncertainty on the calculation of dose. To do this, the Monte Carlo procedure was repeated with the following modifications:

1. The absorption parameters were varied but all other parameters were fixed at their median values.
2. The absorption parameters were fixed at their median values (lognormal distributions) or modal values (triangular distributions) but all other parameters were varied.

3.4.2 Results

3.4.2.1 *Uranium*

Uncertainties on doses from ingestion

Summary statistics of effective dose per unit ingestion of ^{238}U for 1 and 10 year old children and adults are provided in Table 3.1, where they are compared with the ICRP Publication 72 dose coefficients for ingestion by members of the public (ICRP 1996).

The median values of the distributions are reasonably close to the ICRP Publication 72 values (ICRP 1996), but the means of the distributions are around 50% higher than the ICRP values; this is because the model parameters were sampled from broad lognormal distributions which induces a degree of positive skewness into the distribution of dose. The ratio between the 2.5% or 97.5% values and the ICRP reference values is broadly similar for all three age groups, ranging from a factor of 2.4

to 4.6. The 2.5% and 97.5% values are fairly symmetrically distributed (geometrically) about the ICRP values, although the symmetry appears to decrease with age.

In the sensitivity analysis, where only the f_1 parameter value was varied, the effective dose per unit ingestion for adults followed a lognormal distribution with median value equal to the ICRP value; the 2.5% and 97.5% values were within a factor of 3 of the ICRP value. The simulation where the f_1 parameter was fixed at the ICRP value but the systemic parameter values were varied confirmed that the residual variation in dose can be explained by variability in the systemic parameter values. Thus it appears to be uncertainty in the f_1 parameter that makes the greatest single contribution to the variation in dose in the full analysis, accounting for approximately 35% of the total variance in dose.

Uncertainties on doses from inhalation

Summary statistics of effective dose per unit inhalation of ^{238}U for 1 and 10 year old children and adults are provided in Table 3.2, where they are compared with the ICRP Publication 71 dose coefficients for inhalation by members of the public (ICRP 1995a). The doses are compared to the corresponding dose coefficients for “Type M” materials, the recommended solubility class for uranium compounds in the absence of material specific information (ICRP 1995a).

The mean values of the distributions are around a factor of two higher than the ICRP values for all age groups, with the distribution of dose covering a wide range: the 2.5% and 97.5% values are around a factor of 6 lower, and higher, respectively, than the ICRP dose coefficient for each age group; the median values are fairly close to the ICRP values. The additional sensitivity analysis indicated that around 80% of the variance in the distribution was the result of varying the absorption parameters.

Table 3.1. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from ingestion of 1 Bq of ^{238}U ^a

	1 year old infant	10 year old child	Adult
ICRP ^b	12.0	6.8	4.5
Mean	17.9	10.0	6.1
Median	13.7	7.5	4.7
GSD	1.9	2.1	2.0
Q _L	4.9	2.0	1.3
Q _U	54.6	31.1	19
Mean/ICRP	1.5	1.5	1.4
Q_U/ICRP	4.6	4.6	4.2
ICRP/Q_L	2.4	3.4	3.5

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^bICRP Publication 69 value (ICRP 1995b) ($f_1=0.02$).

Table 3.2. Uncertainty analysis of effective dose, E(50) ($\times 10^{-6}$ Sv per Bq) from inhalation of 1 Bq of $^{238}\text{U}^{\text{a}}$

	1 year old infant	10 year old child	Adult
ICRP ^b	9.4	4.0	2.9
Mean	16.3	6.4	4.5
Median	12.4	4.4	3.2
GSD	2.6	2.4	2.5
Q _L	1.5	0.7	0.5
Q _U	57.1	23.8	18
Mean/ICRP	1.7	1.6	1.6
Q_U/ICRP	6.1	5.9	6.1
ICRP/Q_L	6.3	5.7	5.8

^aKey: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^bICRP Publication 71 value (ICRP 1995a) ($f_1=0.02$).

3.4.2.2 Radium

Uncertainties on doses from ingestion

Summary statistics of effective dose per unit ingestion of ^{226}Ra for 1 and 10 year old children and adults are provided in Table 3.3 and compared with the ICRP Publication 67 dose coefficients for members of the public (ICRP 1993).

The factors above and below the ICRP value for the mean, median, 2.5% and 97.5% values are similar for the three age groups. The 97.5% and 2.5% values are a factor of 3-5 greater and less than the ICRP values. The additional sensitivity analysis suggested that the f_1 parameter contributes around 30% of the total variance in dose.

Uncertainties on doses from inhalation

Summary statistics of effective dose per unit inhalation of ^{226}Ra for 1 and 10 year old children and adults are provided in Table 3.4, where they are compared with the ICRP Publication 71 dose coefficients for inhalation of Type S materials by members of the public (ICRP 1995a).

The mean value is around a factor of two greater than the ICRP value for each age group. The distributions of dose also cover a broad geometric range; the upper 97.5% value to ICRP values ranges from a factor of 5 (1 year olds) to a factor of 8 (adults). The ratio of the ICRP values to the 2.5% values range from a factor of 4 (10 year olds) to a factor of 7 (adults); the sensitivity analysis indicated that variation in the absorption parameters constituted around 80% of the total variance, and therefore much of the observed range, in dose.

Table 3.3. Uncertainty analysis of effective dose, E(50) ($\times 10^{-7}$ Sv per Bq) from ingestion of 1 Bq of ^{226}Ra ^a

	1 year old infant	10 year old child	adult
ICRP ^a	9.7	8.1	2.8
Mean	11.1	9.0	3.1
Median	8.5	7.2	2.5
GSD	1.9	2.0	1.9
Q _L	2.6	1.7	0.8
Q _U	32.4	27.5	8.6
Mean/ICRP	1.1	1.1	1.1
Q_U/ICRP	3.4	3.4	3.1
ICRP/Q_L	3.7	4.8	3.5

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 67 value (ICRP 1993)

Table 3.4. Uncertainty analysis of effective dose, E(50) ($\times 10^{-6}$ Sv per Bq) from inhalation of 1 Bq of ^{226}Ra ^a

	1 year old infant	10 year old child	Adult
ICRP ^b	29.0	12.0	9.5
Mean	47.4	22.4	18.9
Median	36.8	14.9	11.2
GSD	2.4	2.5	2.8
Q _L	5.5	2.7	1.4
Q _U	153	84.8	77.2
Mean/ICRP	1.6	1.9	2.0
Q_U/ICRP	5.3	7.1	8.1
ICRP/Q_L	5.3	4.4	6.8

^aKey: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2-5%.

^bICRP Publication 71 values for Type S materials (ICRP 1995a).

3.4.2.3 Caesium

Uncertainties on doses from ingestion

Table 3.5 provides summary statistics of the calculated distributions of effective dose per unit ingestion for all age groups. Uncertainties on doses calculated using the revised model for caesium described by Leggett et al (2003) are also provided (column “Adult (new model)”).

The mean and median values of the distributions of dose calculated using the ICRP Publication 56 model (ICRP 1989) are close to the ICRP Publication 72 dose coefficients (ICRP 1996), for all age groups; the 2.5% and 97.5% values are within a factor of 2 of the dose coefficient. The distribution of dose calculated using the revised model is shifted downwards with respect to the dose coefficient for adults, with the

result that the 97.5% value is close to the value of the dose coefficient; the 2.5% value is a factor of three lower.

3.4.2.4 *Uncertainties on doses from inhalation*

Summary statistics for the calculated distributions of effective dose per unit inhalation for all age groups are given in Table 3.6. Uncertainties on doses calculated using the modified systemic model for caesium described by Leggett et al (2003) are also provided (column "Adult (new model)"). In all cases the mean and median values are close to the values of the dose coefficients for inhalation of caesium given in ICRP Publication 71 (ICRP 1995a). The revised systemic model had little effect on the distribution of dose. The sensitivity analyses indicated that the uncertainty on the values of the dissolution parameters and particle transport clearance had the greatest overall affect on the distribution of dose, accounting for around 50% of the observed variance in the distribution; the dissolution parameters accounted for around 30% of observed variance in the distribution of dose.

Table 3.5. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from ingestion of 1 Bq of ^{137}Cs

	1 year old infant	10 year old child	Adult	Adult (new model)
ICRP ^a	1.2	1.0	1.3	1.3
Mean	1.2	0.94	1.2	0.79
Median	1.2	0.91	1.2	0.76
GSD	1.2	1.4	1.3	1.3
Q _L	0.84	0.48	0.68	0.45
Q _U	1.8	1.5	1.9	1.3
Mean/ICRP	1.0	0.9	0.9	0.6
Q_U/ICRP	1.5	1.5	1.5	1.02
ICRP/Q_L	1.4	2.1	1.9	2.9

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 72 value (ICRP 1996) ($f_1=1.0$).

Table 3.6. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from inhalation of 1 Bq of ^{137}Cs

	1 year old infant	10 year old child	Adult	Adult (new model)
ICRP ^a	2.9	1.3	0.97	0.97
Mean	2.9	1.3	0.97	0.97
Median	2.4	1.1	0.86	0.78
GSD	1.7	1.5	1.6	1.7
Q _L	0.97	0.48	0.38	0.33
Q _U	8.3	3.2	2.3	2.5
Mean/ICRP	1.0	1.0	1.0	1.0
Q_U/ICRP	2.8	2.4	2.4	2.6
ICRP/Q_L	3.0	2.7	2.6	2.9

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) ($f_1=0.1$).

3.4.2.5 Strontium

Ingestion

Summary statistics of effective dose per unit ingestion of ^{90}Sr for 1 and 10 year old children, and adults are provided in Table 3.7, where they are compared with the ICRP Publication 72 dose coefficients for ingestion by members of the public (ICRP 1996).

The mean and median values of the distributions are close to the ICRP values for all age groups. The geometric distribution of the 2.5% and 97.5% values is fairly symmetric around the ICRP values, being typically around a factor of 2-4, and reasonably consistent between age groups. The sensitivity analysis indicated that variation in the f_1 parameter contributes around 20% of the total variance in dose.

Inhalation

Summary statistics of effective dose per unit inhalation of ^{90}Sr for 1 and 10 year old children and adults are provided in Table 3.8 where they are compared with the ICRP Publication 71 dose coefficients for inhalation by members of the public (ICRP 1995a). For strontium, comparisons are made with the dose coefficients derived for “Type M” materials, which is the default ICRP recommendation for environmental forms of strontium compounds.

The mean and median values of dose are a factor of 1.3-1.6 higher than the ICRP values. The 2.5% and 97.5% values of the distributions are around a factor of 2 (10 year olds) to 3 (adults) lower, and a factor of 3 (adults) to 4 (1 year olds) higher, respectively, than the ICRP values. The additional analysis indicated that variation in the absorption parameters contributes a relatively small proportion of the total variation in dose (15%).

Table 3.7. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from ingestion of 1 Bq of ^{90}Sr

	1 year old infant	10 year old child	Adult
ICRP ^a	7.3	6.0	2.8
Mean	7.3	5.8	2.9
Median	6.3	5.1	2.4
GSD	1.6	1.8	1.8
Q _L	2.7	1.7	0.8
Q _U	17.2	14.0	7.4
Mean/ICRP	1.0	1.0	1.0
Q_U/ICRP	2.4	2.3	2.6
ICRP/Q_L	2.7	3.6	3.6

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 72 value (ICRP 1996) ($f_1=1.0$).

Table 3.8. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from inhalation of 1 Bq of ^{90}Sr

	1 year old infant	10 year old child	Adult
ICRP ^a	11.0	5.1	3.6
Mean	15.5	8.1	4.8
Median	13.6	7.4	4.3
GSD	1.7	1.6	1.7
Q _L	4.6	2.9	1.4
Q _U	39.5	18.0	11.8
Mean/ICRP	1.4	1.6	1.3
Q_U/ICRP	3.6	3.5	3.3
ICRP/Q_L	2.4	1.8	2.6

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) ($f_1=0.1$).

3.4.2.6 Plutonium

Uncertainties on doses from ingestion

Summary statistics of the distributions of effective dose per unit ingestion of ^{239}Pu for 1 and 10 year old children and adults are provided in Table 3.9, and compared to the ICRP Publication 72 dose coefficients for members of the public (ICRP 1996). The distributions of dose for adults calculated by Puncher and Harrison (2012b) using the revised model are also provided.

The factors above and below the ICRP value for the mean, median, 2.5% and 97.5% values are similar for the three age groups; mean values are around 10-20% higher than the ICRP values; median values are close to the ICRP values. The 97.5% and 2.5% values are a factor of 3 greater and less than the ICRP values. The sensitivity

analysis showed that nearly all of the uncertainty on effective dose was accounted for by uncertainty on the f_1 value.

Uncertainties on doses from inhalation

Summary statistics of the distributions of effective dose per unit inhalation of ^{239}Pu for 1 and 10 year old children, and adults are provided in Table 3.10; they are compared with the ICRP Publication 71 dose coefficients for inhalation of Type S materials by members of the public (ICRP 1995a).

The mean and median values are around a factor of two to three greater than the ICRP value for all age groups. The distributions of dose also cover a broad geometric range; the ratio of the 97.5% value to ICRP values ranges from a factor of 6 (1 year olds) to a factor of 8 (adults). The ratio of the ICRP values to the 2.5% values is around a factor of two for all age groups.

Table 3.9. Uncertainty analysis of effective dose, E(50) ($\times 10^{-7}$ Sv per Bq) from ingestion of 1 Bq of ^{239}Pu

	1 year old infant	10 year old child	Adult	Adult (new model) ^a
ICRP ^a	4.2	2.7	2.5	2.5
Mean	4.8	3.1	2.9	3.0
Median	4.2	2.7	2.5	2.5
GSD	1.7	1.7	1.7	1.7
Q _L	1.6	0.88	0.84	0.86
Q _U	11.7	7.8	7.3	7.8
Mean/ICRP	1.1	1.1	1.2	1.2
Q_U/ICRP	2.8	2.9	2.9	2.9
ICRP/Q_L	2.7	3.1	3.0	3.0

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aFrom Puncher and Harrison (2012b)

^bICRP Publication 72 value (ICRP 1996) ($f_1=0.0005$).

Table 3.10. Uncertainty analysis of effective dose, E(50) ($\times 10^{-5}$ Sv per Bq) from inhalation of 1 Bq of ^{239}Pu

	1 year old infant	10 year old child	Adult	Adult (new model)
ICRP ^a	3.9	1.9	1.6	1.6
Mean	9.4	5.3	4.9	4.9
Median	8.0	4.7	4.3	4.4
GSD	1.9	1.9	2.0	2.0
Q _L	2.3	1.0	0.79	0.87
Q _U	24.6	12.4	12.5	12.8
Mean/ICRP	2.4	2.8	3.1	3.1
Q_U/ICRP	6.3	6.5	7.8	8.0
ICRP/Q_L	1.7	1.9	2.0	1.8

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) ($f_1=0.00001$).

3.4.2.7 Americium

Uncertainties on doses from ingestion

Summary statistics of the distributions of effective dose per unit ingestion of ^{241}Am for 1 and 10 year old children and adults are provided in Table 3.11, and compared with the ICRP Publication 72 dose coefficients for members of the public (ICRP 1996).

Table 3.11. Uncertainty analysis of effective dose, E(50) ($\times 10^{-7}$ Sv per Bq) from ingestion of 1 Bq of ^{241}Am

	1 year old infant	10 year old child	Adult ^a
ICRP ^b	3.7	2.2	2.0
Mean	4.2	2.5	2.3
Median	3.6	2.1	2.0
GSD	1.7	1.8	1.8
Q _L	1.4	0.70	0.63
Q _U	11.0	6.6	6.1
Mean/ICRP	1.1	1.1	1.2
Q_U/ICRP	3.0	3.0	3.1
ICRP/Q_L	2.7	3.3	3.2

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aFrom Puncher and Harrison (2012b)

^bICRP Publication 72 value (ICRP 1996) ($f_1=0.0005$).

The factors above and below the ICRP value for the the mean, median, 2.5% and 97.5% values are similar for the three age groups; mean values are around 10-20% higher than the ICRP values; median values are close to the ICRP values. The 97.5% and 2.5% values are a factor of 3 greater and less than the ICRP values. Additional sensitivity analysis indicated that nearly all the uncertainty on dose was accounted for by uncertainty on the f_1 value.

Uncertainties on doses from inhalation

Summary statistics of effective dose per unit inhalation of ²⁴¹Am for 1 and 10 year old children and adults are provided in Table 3.12 where they are compared with the ICRP Publication 71 dose coefficients for inhalation of Type S materials by members of the public (ICRP 1995a).

The mean and median values are around a factor of two to three greater than the ICRP value for each age group. The distributions of dose also cover a broad geometric range; the upper 97.5% value to ICRP values ranges from a factor of 6 (1 year olds) to a factor of 8 (adults). The ratio of the ICRP values to the 2.5% values is around a factor of two for all age groups.

Table 3.12. Uncertainty analysis of effective dose, E(50) (x10⁻⁵ Sv per Bq) from inhalation of 1 Bq of ²⁴¹Am

	1 year old infant	10 year old child	Adult
ICRP ^a	4.0	1.9	1.6
Mean	9.3	5.0	4.8
Median	8.2	4.4	4.1
GSD	1.8	1.9	2.0
Q _L	2.6	1.1	0.81
Q _U	22.6	12.3	12.4
Mean/ICRP	2.3	2.6	3.0
Q_U/ICRP	5.7	6.5	7.8
ICRP/Q_L	1.5	1.8	2.0

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) (f₁=0.0005).

3.4.2.8 Tritium

Uncertainties on doses from ingestion or inhalation of tritiated water (HTO)

Summary statistics of the distributions of effective dose per unit ingestion/inhalation of ³H as tritiated water (HTO) for 1 and 10 year old children, and adults are provided in Table 3.13; they are compared with the corresponding ICRP Publication 72 dose coefficients for members of the public (ICRP 1996). The mean and median values of the distributions are close to the ICRP values, with the 2.5% and 97.5% values being within a factor of two of the ICRP dose coefficients for all age groups.

Uncertainties on doses from ingestion of organically bound tritium (OBT)

Summary statistics of the distributions of effective dose per unit ingestion/inhalation of ³H as organically bound tritium (OBT) for 1 and 10 year old children and adults are provided in Table 3.14. They are compared with the corresponding ICRP Publication 72 dose coefficients for members of the public (ICRP 1996). The mean and median values of the distributions are close to the ICRP values, with the 2.5% and 97.5% values being around a factor of two of the ICRP dose coefficients for all age groups.

Table 3.13. Uncertainty analysis of effective dose, E(50) ($\times 10^{-11}$ Sv per Bq) from inhalation or ingestion of 1 Bq of HTO

	1 year old infant	10 year old child	Adult
ICRP ^a	4.8	2.3	1.8
Mean	5.4	2.5	2.0
Median	5.3	2.5	2.0
GSD	1.2	1.2	1.2
Q _L	3.7	1.7	1.4
Q _U	7.7	3.7	2.9
Mean/ICRP	1.1	1.1	1.1
Q_U/ICRP	1.6	1.6	1.6
ICRP/Q_L	1.3	1.3	1.3

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 72 value (ICRP 1996).

Table 3.14. Uncertainty analysis of effective dose, E(50) ($\times 10^{-11}$ Sv per Bq) from ingestion of 1 Bq of OBT

	1 year old infant	10 year old child	Adult
ICRP ^a	12.0	5.7	4.2
Mean	12.3	5.7	4.2
Median	11.0	5.0	3.7
GSD	1.5	1.5	1.5
Q _L	6.2	2.7	2.0
Q _U	25.6	12.2	9.2
Mean/ICRP	1.0	1.0	1.0
Q_U/ICRP	2.1	2.1	2.2
ICRP/Q_L	1.9	2.1	2.1

^aKey: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^bICRP Publication 72 value (ICRP 1996).

3.4.2.9 Iodine

Uncertainties on doses from ingestion

Summary statistics of effective dose per unit ingestion of ¹³¹I and ¹²⁹I for 1 and 10 year old children and adults are provided in Tables 3.15 and 3.16, respectively, where they are compared with the corresponding ICRP Publication 72 dose coefficients for members of the public (ICRP 1996). For ingestion of ¹³¹I, the mean values are around a factor of 1.5 higher than the ICRP value; the 2.5% values are equal to the ICRP values and the 97.5% values are a factor of two higher than the ICRP values. Assuming the ICRP Publication 56 model or the revised model proposed by Leggett (2010) appears to make little difference to the calculated distribution of dose.

For ^{129}I , the mean and median values of the distributions of dose calculated using the ICRP Publication 56 model are around a factor of two greater than the ICRP values; the 2.5% values are 10% to 20% higher than the ICRP values; the 97.5% values are around a factor of 3.5 higher than the ICRP values. The distribution of dose per unit ingestion calculated using the revised model for adults is shifted slightly upwards with respect to the distribution calculated using the ICRP Publication 56 model: the mean value is a factor of 2.4 higher than the ICRP value; the 2.5% value is equal to the ICRP value, and the 97.5% value is a factor of 4 greater than the ICRP value.

Uncertainties on doses from inhalation

Summary statistics of effective dose per unit inhalation of ^{131}I and ^{129}I , for 1 and 10 year old children and adults are provided in Tables 3.17 and 3.18, respectively. They are compared with the corresponding ICRP Publication 71 dose coefficients for members of the public for inhalation of methyl iodide vapour (ICRP 1995a). For inhalation of ^{131}I , the mean values are around a factor of 1.3 - 1.4 higher than the ICRP values; the ICRP values are a factor of 1.4-1.5 higher than the 2.5% values; the 97.5% values are a factor of 2 greater than the ICRP values. The distributions of effective dose per unit inhalation for adults are similar whether calculated using the ICRP Publication 56 model or the revised model for iodine proposed by Leggett (2010).

For ^{129}I , the mean values of the distributions of dose calculated using the ICRP Publication 56 model are around a factor of 1.6-1.7 higher than the ICRP values; the ICRP values are around a factor of 1.4-1.5 higher than the 2.5% values; the 97.5% values range from a factor of 3.1 (10 year olds) to 3.3 (adults) higher than the ICRP values. The distribution of dose per unit inhalation calculated using the revised model for adults is shifted slightly upwards with respect to the distribution calculated using the ICRP Publication 56 model, although the effect appears to be less pronounced than that observed for ingestion. The mean value is a factor of 1.9 greater than the ICRP value; the ICRP value is around a factor of 1.3 higher than the 2.5% value, and the 97.5% value a factor of 3.5 higher than the ICRP value. The difference between the distributions of dose for inhalation calculated using the revised systemic model and the ICRP Publication 56 model is less pronounced than those for ingestion, presumably due to the dominant contribution of the vapour deposition parameter, V_{DP} , in determining the amount of iodine that is deposited in the lung and available for uptake to blood and subsequent uptake to the thyroid.

Table 3.15. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from ingestion of 1 Bq of ^{131}I ^a

	1 year old infant	10 year old child	Adult	Adult (new model)
<i>ICRP</i> ^a	18.0	5.2	2.2	2.2
Mean	28.4	8.0	3.3	3.5
Median	28.3	8.0	3.3	3.5
GSD	1.2	1.2	1.2	1.2
Q _L	17.6	5.3	2.1	2.3
Q _U	40.6	11.0	4.6	4.7
Mean/ICRP	1.6	1.5	1.5	1.6
Q_U/ICRP	2.3	2.1	2.1	2.1
ICRP/Q_L	1.02	0.98	1.05	0.96

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 72 value (ICRP 1996) ($f_1=1$).

Table 3.16. Uncertainty analysis of effective dose, E(50) ($\times 10^{-7}$ Sv per Bq) from ingestion of 1 Bq of ^{129}I

	1 year old infant	10 year old child	Adult	Adult (new model)
<i>ICRP</i> ^a	2.2	1.9	1.1	1.1
Mean	4.1	3.6	2.0	2.4
Median	3.9	3.4	1.9	2.2
GSD	1.4	1.4	1.4	1.4
Q _L	1.9	1.7	1.0	1.1
Q _U	7.4	6.7	3.6	4.4
Mean/ICRP	1.9	1.9	1.8	2.2
Q_U/ICRP	3.4	3.5	3.3	4.0
ICRP/Q_L	1.2	1.1	1.1	1.0

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 72 value (ICRP 1996) ($f_1=1$).

Table 3.17. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from inhalation of 1 Bq of ^{131}I

	1 year old infant	10 year old child	Adult	Adult (new model)
ICRP ^a	13	3.7	1.5	1.5
Mean	17	4.9	2.0	2.1
Median	17	4.8	2.0	2.0
GSD	1.3	1.3	1.3	1.3
Q _L	8.7	2.6	1.1	1.1
Q _U	26	7.3	2.9	3.0
Mean/ICRP	1.3	1.3	1.3	1.4
Q_U/ICRP	2.0	2.0	1.9	2.0
ICRP/Q_L	1.5	1.4	1.4	1.4

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) ($f_1=1$).

Table 3.18. Uncertainty analysis of effective dose, E(50) ($\times 10^{-8}$ Sv per Bq) from inhalation of 1 Bq of ^{129}I

	1 year old infant	10 year old child	Adult	Adult (new model)
ICRP ^a	15	13	7.4	7.4
Mean	24.6	21.5	12.1	13.8
Median	23.0	20.5	11.6	13.0
GSD	1.5	1.5	1.5	1.5
Q _L	11.0	8.8	5.1	5.9
Q _U	48.0	40.8	24.5	25.7
Mean/ICRP	1.6	1.7	1.6	1.9
Q_U/ICRP	3.2	3.1	3.3	3.5
ICRP/Q_L	1.4	1.5	1.5	1.3

Key: GSD – geometric standard deviation; Q_U – the upper 97.5 percentile; Q_L – the lower 2.5 percentile. Statistics represent the arithmetic mean of three simulations; the coefficient of variation of the simulations (the standard deviation of the mean of the three runs over the mean) was within 2%.

^aICRP Publication 71 value (ICRP 1995a) ($f_1=1$).

3.4.3 Uncertainty analysis - discussion

3.4.3.1 Uranium

Ingestion

The analysis indicates that uncertainty on the value of the f_1 parameter makes the greatest single contribution to the uncertainty on dose resulting from ingestion of uranium. When only this parameter was varied in the sensitivity analysis, the ratio of the 97.5% value to dose coefficient suggests that the uncertainty on the location of the mean value of dose, with respect to the published value, is around a factor of 2.4 for adults. The same analysis conducted for 1 and 10 year old children gave a higher value of 2.7, presumably due to the slightly wider range of f_1 values assumed for these age groups. It is assumed that the distributions derived for the systemic parameters reflect mainly variability in the retention and excretion of uranium from the body, although it is

acknowledged that the location (mean value) of the distributions is also uncertain. Consequently, an uncertainty factor of 4.2, suggested by the ratio of 97.5% value:ICRP value in the full uncertainty analysis, is probably a conservative estimate of the uncertainty on the location of the mean.

Uncertainty factors for ingestion of ^{238}U

Because the mean of the distribution of dose is only 20% higher than the ICRP value in simulation where only the systemic parameters were varied, it seems reasonable to suppose that the mean is more likely to be around a factor of 2-3 above the ICRP value rather than a factor of 4-5. On this basis it is concluded that the UF for effective dose per unit ingestion of ^{238}U , is within a factor of 3 for all three age groups.

Inhalation

For inhalation of uranium, the results show that it is the distributions derived for the absorption parameters that have the greatest impact on the variance of the distribution of dose. Because this distribution represents a mixture of uranium bearing materials, a factor of 6 (the ratio between the 2.5% or 97.5% values and the ICRP values in Table 3.2) is a conservative estimate of the uncertainty on the location of the mean value of effective dose per unit intake with respect to the ICRP values. Instead, the ratio difference between the means of the distributions and the ICRP values is more indicative of the uncertainty: the ratio between the mean and the ICRP value is 1.6-1.7.

Uncertainty factors for inhalation of ^{238}U

It is concluded that the UF for effective dose per unit inhalation of environmental forms of ^{238}U is around a factor of 2 for all age groups, with a factor of 6 being very much a conservative upper limit.

3.4.3.2 Radium

Ingestion

The analysis indicates that variation (variability) in the systemic parameters, rather than the f_1 value, accounts for much of the observed variation in effective dose following ingestion of ^{226}Ra . The assumption is made that this applies to all age groups.

Uncertainty factors for ingestion of ^{226}Ra

Given that variability in systemic retention of radium accounts for a significant proportion of the observed factor of 3-4 fold variation in effective dose per unit ingestion, as indicated by the ratios between the calculated 97.5% or 2.5% values and the ICRP values, it is concluded that the UF for the ICRP dose coefficients for ingestion of ^{226}Ra for all age groups is around a factor of 3.

Inhalation

The analysis suggests that the wide range of doses observed for radium is primarily caused by the distributions derived for the absorption parameters. When only these were varied the 2.5% value was around a factor of 6 lower than the ICRP dose coefficient for ^{226}Ra (Type S compounds), the 97.5% value a factor of 4 higher. In this analysis it is assumed that variation in these parameters mainly reflects uncertainty on the lung solubility of radium present in vitrified fly ash. Additional calculations showed that the effect of the absorption parameters is exacerbated by the revised HRTM

structure, which predicts higher lung doses than the existing HRTM for low solubility compounds (Birchall et al 2010). It was found that the effective dose per unit inhalation of ^{226}Ra calculated using the revised structure is $18.5 \times 10^{-6} \text{ Sv Bq}^{-1}$ for adults, assuming Type S absorption parameters and ICRP Publication 71 deposition and breathing parameters (i.e. ICRP parameter values for environmental ^{226}Ra bearing aerosols), compared with the current ICRP value of $9.5 \times 10^{-6} \text{ Sv Bq}^{-1}$. This explains why the median value of dose from the simulation where the absorption values were fixed at their median values has a value of $18.3 \times 10^{-6} \text{ Sv Bq}^{-1}$ rather than a value closer to $9.5 \times 10^{-6} \text{ Sv Bq}^{-1}$. It seems reasonable to suppose that the simulation where only the absorption parameters were varied gives a more indicative range of the uncertainty on the mean value of dose than does the simulation where all parameters were varied. The results suggests that the mean values are within a factor of 6 of the ICRP reference values for Type S compounds for all age groups, with the ratio between the 97.5% value and ICRP value being closer to 4.

Uncertainty factors for inhalation of ^{226}Ra

Based on the above, it is inferred that the UF for the ICRP dose coefficients for inhalation of environmental aerosols bearing ^{226}Ra for all age groups is around a factor of 6. Note that the revised HRTM particle transport clearance model will be used by ICRP to calculate revised dose coefficients for members of the public. This will likely reduce the ratio between the 97.5% value of the distribution and the new coefficient value to around two. The analysis also indicates that better data describing the dissolution and uptake to blood of radium present in fly ash will significantly reduce uncertainties on doses from inhalation of ^{226}Ra present in this form.

3.4.3.3 *Caesium*

Ingestion

Uncertainties on dose per unit ingestion of ^{137}Cs for adults have been calculated previously by Apostoaei and Miller (2004), who conducted a parameter uncertainty analysis based on the ICRP Publication 56 model for caesium (ICRP 1989). Although the authors calculated distributions of weighted equivalent organ dose, instead of committed effective dose, the median values were close to the published values for different tissues, with geometric standard deviations ranging between 1.2 and 1.5, which is consistent with the results observed here: the mean and median values of effective dose per unit ingestion are close to the ICRP values for all age groups, with GSDs of around 1.2-1.4, when calculated using the ICRP Publication 56 model. This suggests that the distribution of effective dose is fairly insensitive to the assumption that caesium retention is correlated with bodily potassium content, which was assumed by Apostoaei and Miller (2004). The distribution is also relatively insensitive to the assumed model structure. There are notable conceptual differences between the revised model for caesium and its more empirically based predecessor; nevertheless, the parameter uncertainty analyses conducted with both models produce similar distributions of effective dose per unit ingestion. This is perhaps not surprising since both models produce similar distributions of whole body retention of ^{137}Cs at different times after an acute intake. The small differences in committed effective dose arise because of small differences in the temporal distribution of caesium to systemic organs that are assigned ICRP Publication 60 tissue weighting factors.

Uncertainty factors for ingestion of ^{137}Cs

The results suggest that the UF for ingestion of ^{137}Cs is within a factor of 3 for all age groups; this may be slightly conservative because the distributions of dose produced using either model are dominated by parameter variability.

Inhalation

The results suggest that the uncertainty on dose per unit inhalation of ^{137}Cs is insensitive to the structure of the systemic model, and is a consequence of the effect noted for ingestion – both models predict similar whole body retention of caesium - and also because the weighted equivalent dose to the lung constitutes around 50% of the effective dose (calculated assuming the modal value for the distribution of f_r , and the median value for s_b). The dispersion of the distribution of dose is determined mostly by uncertainty on the lung solubility of the caesium bearing aerosol and variability in particle transport clearance. When only the absorption parameters were varied, the 2.5% and 97.5% values of the distribution of dose were within a factor of two of the dose coefficient.

Uncertainty factors for inhalation of ^{137}Cs

Overall, the results suggest that the UF for inhalation of ^{137}Cs is around a factor of 2-3 for all age groups.

3.4.3.4 Strontium

Ingestion

The results of the uncertainty analysis for ingestion of strontium are reasonably consistent with the previous study by Apostoaei and Miller (2004), who applied the method to calculate ranges of equivalent organ doses, but not effective dose, for adult males and females following ingestion of ^{90}Sr , using the ICRP Publication 67 systemic model for strontium that was also used in this study. Their analysis gave median values and ranges of equivalent doses to soft tissues such as liver and muscle that were identical to those calculated in the present study; doses to red bone marrow (RBM) and bone surfaces gave identical median values but the 95% range of values were wider in their analysis: equivalent dose to RBM ($\times 10^{-7}$ Sv Bq $^{-1}$) ranged from 0.28 to 60.0, versus 0.4 to 50.4 in this study; equivalent doses to bone surfaces ($\times 10^{-7}$ Sv Bq $^{-1}$) ranged from 0.56 to 19.0, versus 1.0 to 10.5 in this study. From this, and considering the different contributions of the weighted equivalent doses to these organs to effective dose, it is inferred that the analysis of Apostoaei and Miller (2004) would yield distributions of effective dose per unit intake for adult males that are similar to those calculated for adults in the present analysis. A more precise comparison of the two studies is difficult because Apostoaei and Miller (2004) do not state which parameters were varied in their analysis, other than that distributions were derived based on a literature review of strontium metabolism in humans and that selected distributions were adjusted to give ranges of bioassay predictions for whole body, soft tissue and excreta that were consistent with the variation observed in the human data, which is essentially the same as the approach adopted here.

Uncertainty factors for ingestion of ^{90}Sr

The analysis indicates that variation in the systemic parameters (variability), rather than variation in the f_1 value (uncertainty), accounts for much of the observed variation in effective dose following ingestion of ^{90}Sr . Given that variability in systemic retention of strontium accounts for a significant proportion of the observed 3-4 fold variation in effective dose per unit ingestion, as indicated by the ratios between the calculated 97.5% or 2.5% values and the ICRP values, it is concluded that the UF for the ICRP dose coefficients for ingestion of ^{90}Sr for all age groups is closer to a factor of 3.

Inhalation

The results indicate that variation in the absorption parameters have much less of an effect on the distribution of dose compared with some of the other radionuclides, particularly ^{226}Ra . Also, the means of the distributions are closer to the ICRP values than is observed for radium, which is consistent with previous studies that show the revised particle transport model gives similar lung doses to the existing HRTM for soluble aerosols (Type F and M materials) (Birchall et al 2010, Puncher et al 2011) whose rate of clearance from the alveolar-interstitial (AI) region of the lung is not determined by the slower rate of particle transport clearance from this region in the revised model. Additional uncertainty analyses suggested that most of the variation in dose is due to the combined effects of variation in the rate of particle transport clearance and absorption. The former represents primarily variability in the rate of particle transport clearance in the population; the latter uncertainty in the solubility of environmental strontium bearing aerosols in the lung. Thus it is likely that the factor of 2-4 fold variation of the 2.5% and 97.5% values around the ICRP dose coefficients for Type M ^{90}Sr are conservative estimates of the mean value of dose.

Uncertainty factors for inhalation of ^{90}Sr

It is concluded that the UF for inhalation of ^{90}Sr is around a factor of three, for all age groups.

3.4.3.5 *Plutonium and americium*

Ingestion

The sensitivity analysis, where only the f_1 parameter was varied, indicates that nearly all of the uncertainty on effective dose per unit ingestion of plutonium or americium is accounted for by uncertainty on the amount of activity taken up to blood from the alimentary tract. The results also suggest that the uncertainty on dose is relatively insensitive to changes in model structure. Assuming either the ICRP Publication 67 model for plutonium or the revised model described by Leggett et al (2005) has little effect on the calculated distribution of dose per unit ingestion for plutonium.

Uncertainty factors for ingestion of ^{239}Pu and ^{241}Am

The results suggest a UF of 3 for plutonium and americium for all age groups, consistent with the previous study by Puncher and Harrison (2012b) who calculated uncertainties on doses for adults resulting from ingestion of plutonium and americium.

Inhalation

There is an upward shift in the distribution of dose per unit inhalation of ^{239}Pu and ^{241}Am with respect to the published dose coefficients. This asymmetry is a consequence of the revised particle transport model used in the uncertainty analysis, which predicts higher lung doses for low solubility alpha emitters. The distributions of dose for plutonium were similar whether the uncertainty analysis was performed using the ICRP Publication 67 model or revised systemic model described by Leggett et al (2005). This occurs partly because effective dose per unit inhalation of insoluble plutonium is mainly determined by the weighted equivalent dose to the lung; and also partly because the effective dose per unit injection, where doses to systemic tissues dominate the calculation of effective dose, are similar when calculated using either model ($5.07 \times 10^{-4} \text{ Sv Bq}^{-1}$ versus $4.93 \times 10^{-4} \text{ Sv Bq}^{-1}$, for the revised and ICRP Publication 67 models, respectively).

Surprisingly, additional runs indicated that the wide range in dose was the result of variation in particle transport clearance rather than uncertainty on the dissolution parameters, which, when varied alone produced a small variation in dose per unit inhalation for adults, albeit with doses shifted to higher values. For plutonium, the range of 2.5% to 97.5% values was 3.1×10^{-5} to $3.7 \times 10^{-5} \text{ Sv Bq}^{-1}$; or a factor of 1.9-2.3 higher than the dose coefficient for insoluble (Type S) plutonium; and for americium, the range of 2.5% to 97.5% values was 3.0×10^{-5} to $4.1 \times 10^{-5} \text{ Sv Bq}^{-1}$; or a factor of 1.9-2.6 higher than the dose coefficient for insoluble (Type S) americium. The small range is because when values of the dissolution parameters at the higher end of the derived distributions are assumed (the material is more soluble) the effective dose per unit inhalation of plutonium or americium is dominated by the large contribution to effective dose from the weighted equivalent dose to systemic organs; in particular, doses to liver, bone surfaces and red bone marrow. These results are consistent with the fact that the effective dose coefficient per unit inhalation is around a factor of three higher for soluble plutonium or americium (Type M material) compared with insoluble plutonium or americium (Type S material) (ICRP 1995a).

Uncertainty factors for inhalation of ^{239}Pu and ^{241}Am

The results suggest that the UF for inhalation of plutonium or americium is likely to be closer to 3 (all age groups), than 6 (1 and 10 year olds) or 8 (adults).

3.4.3.6 Tritium

The present analysis applies the conventional one stage Monte Carlo method to estimate uncertainty on the population mean value of effective dose per unit intake of tritiated water (HTO) and dietary organically bound tritium (OBT) by propagating uncertainty on the mean values of biokinetic parameters into the distribution of dose. As noted in section 2, a correct approach to estimate uncertainty on the mean in a non-linear system is to use a two stage Monte Carlo method that differentiates the effects of parameter uncertainty and variability in estimating uncertainty on the population mean. Use of the one stage Monte Carlo method may therefore introduce a bias in this analysis. However, it was found that using the distributions derived by Harrison et al (2002), which are wider than the distributions assumed here (discussed in Appendix C4.6), gave distributions of effective dose per unit intake of HTO, for example, that:

A. Contained the distribution calculated here: the 95% range of values was 1.25×10^{-11} - 4.68×10^{-11} Sv Bq⁻¹, versus 1.4×10^{-11} - 2.9×10^{-11} Sv Bq⁻¹ calculated here.

B. Have a slightly higher mean value of 2.3×10^{-11} Sv Bq⁻¹ versus 2.0×10^{-11} Sv Bq⁻¹.

These results can be explained by the higher variance and greater degree of positive skewness of the distributions derived by Harrison et al (2002) compared with those used in the present analysis. This suggests that any bias introduced by using the single stage Monte Carlo would be small, and limited to a small positive shift in the distribution of the mean dose per unit intake caused by the lognormal distributions representing variability in the rate of loss of activity from T₁ and T₂ (note that the variance term representing variability is integrated out, to give a mean value, in the two stage Monte Carlo).

A more significant source of bias might be the effect of uncertainty in model structure. The current models for HTO and OBT are simple compartment models intended for radiation protection purposes; but are they too simple? Although the short term compartment probably adequately represents the turnover of body water, the long term compartment is a simplistic representation of the distribution and clearance of tritium incorporated into dietary organic material. In reality, organically bound tritium follows closely the metabolism of carbon; the rate of turnover depends on the chemical form in which it is incorporated. For example, Taylor (2004) compared doses from ¹⁴C labelled materials using material specific half-lives, with the dose calculated from intakes of ¹⁴C using the model for carbon assumed by ICRP (1989), and found that although a number of compounds gave doses close to the ICRP value, for others the ratio between the calculated dose and the ICRP value ranged from 5 to 200.

Several authors have proposed alternative models that describe physiologically more realistic pathways of tritium retention and clearance by the body. Whillans (2003) describes a more physiological model for carbon/organically bound tritium that is based on the rate of turnover of carbon in carbohydrate, fat and protein pools of the body. Development of the model was motivated by the requirement for a model that gave realistic predictions of ¹⁴C excretion in urine following inhalation of ¹⁴C for retrospective dosimetry (doses estimated from urine or other bioassay measurements), rather than to obtain better estimates of dose per unit inhalation. Whillans (2003) does not provide dose estimates using the proposed model.

Richardson and Dunford (2003a; 2003b) proposed two biochemically based models for estimating doses from dietary intakes of organically bound tritium; the two models have a similar basis but differ in complexity. For both models, the input of tritium is a function of the amount of tritium present in carbohydrate, fat and protein, and the quantities of these materials consumed in a typical diet. In the simpler model, tritium was distributed to carbohydrate, fat and protein pools. The complex model included these pools, but also included additional compartments to model long term retention of tritium in glycogen, adipose tissue and structural protein components in bone and connective tissue. Given the level of complexity, the model predicts estimates of effective dose per unit ingestion of HTO and OBT that are in good agreement with the ICRP values; effective dose per unit ingestion for HTO and OBT are a factor of 1.2 and 1.8 higher, respectively, than the doses predicted by the ICRP Publication 56 models for adults.

Galeriu and Melintescu (2010) proposed a metabolic model where tritium retention in body tissues, including brain, soft tissue, muscle and adipose tissue, was determined by the metabolic rate of each organ. The authors adapted the model for different age groups and genders. The proposed model predicts effective doses per unit ingestion that are remarkably close to the ICRP values: doses from HTO were 10-12% higher than the ICRP value for 1 year olds, 10 year olds and adults; doses from OBT were 10-15% higher than the ICRP values for the same age groups.

ICRP is considering revised models for HTO and OBT in its forthcoming publication on occupational intakes of radionuclides (ICRP, in preparation). The models include extra-vascular HTO compartments, and short and long term OBT compartments; there is recycling of activity between the organically bound compartments and the extra-vascular HTO compartment. The models are based on recycling models for HTO proposed by several authors (NCRP 1979; Saito 1992; Hill and Johnson 1993). The models predict effective doses per unit ingestion for adults that are a factor of 1.05 and 1.3 higher than the ICRP Publication 56 dose coefficients for HTO and OBT, respectively, as calculated by the present author using the ICRP Publication 60 tissue weighting factors.

That alternative, and structurally more complex, models give effective doses that are in good agreement with the ICRP values suggests that, although simple, the structure of the ICRP Publication 56 model represents the distribution and retention of tritium in body water and organically bound compartments to an extent that is probably sufficient for prospective radiation protection of population groups. Further, the distributions used in the parameter uncertainty analysis described here provide a reasonable range for the mean values of parameters that determine the mean effective dose per unit intake of environmental forms of tritium: OBT in food, and HTO in water as liquid or vapour.

Uncertainty factors for inhalation of HTO and OBT

Based on the above it is concluded that the UF, considering biokinetic uncertainties, for intakes of tritium in the form of HTO or OBT is around a factor of 2 for all age groups.

3.4.3.7 Iodine

The distributions of effective dose per unit ingestion or inhalation are shifted upwards with respect to the ICRP value for all age groups and this is because the mean 24 hour fractional uptake of iodine by the thyroid, the U_{24} value, is assumed to be higher than the ICRP reference value for a UK population. The distribution of dose also appears to be relatively insensitive to differences in model structure, which is likely to be because both the ICRP Publication 56 model and the revised model for iodine, described by Leggett (2010), broadly predict similar thyroid retention over time. The uncertainty on effective dose per unit intake are higher for intakes of ^{129}I compared with ^{131}I and is because thyroid doses from the former are dependent on the rate of removal of iodine from the thyroid in addition to the rate of uptake. The slight upward shift in effective dose per unit intake of ^{129}I observed with the revised model is because the revision predicts a slightly lower rate (12%) of removal of iodine from the thyroid compared with the ICRP Publication 56 model.

As for tritium, the distributions derived for the iodine analysis reflect uncertainty on the mean values of systemic model parameters (for the lung parameters, only the fraction of inhaled material deposited in the airways is important and variation in this parameter value is linearly related to effective dose), and the calculations were performed using the conventional one stage Monte Carlo method. Thus, the distributions of dose, which are assumed to be estimates of the uncertainty on the mean dose per unit intake, may also be biased. However, it was found that increasing the variance of the lognormal distributions affecting uptake to, and loss from, the thyroid produced a small increase in the variance of the distribution of dose and a small increase in its mean value; the latter can be explained by the positive skewness of the lognormal distribution. This result is consistent with parameter uncertainty analyses of dose per unit ingestion of ^{131}I reported by others, including Dunning and Schwartz (1981) and later by Apostoaei and Miller (2004), and Fritsch (2007). These analyses considered mainly variability in the values of model parameters governing uptake and loss of iodine from the thyroid, but gave distributions of doses with mean values close to the ICRP value, albeit with higher variance than observed here. This suggests that any bias introduced by using a one stage, rather than two stage, Monte Carlo would be small in this case.

Uncertainty factors for ingestion or inhalation ^{131}I and ^{129}I

Based on the above it is concluded that the UF for a UK population, considering biokinetic uncertainties, for ingestion or inhalation of ^{131}I and ^{129}I is around a factor of 2 and around a factor of 3-4, respectively, for all age groups.

4 DISCUSSION

To ensure that exposures by members of the public and workers to sources of ionising radiation are within acceptable limits and constraints, ICRP has provided reference dose coefficients and nominal risk coefficients for populations of “adult workers” and for the “whole population”. In order to assess the reliability of a particular dose coefficient as a protection device for a defined exposure pathway and population group, a useful approach is to compare a best estimate of the detriment adjusted cancer risk coefficient for the exposure with the nominal detriment associated with the effective dose coefficient of interest. A dose coefficient can be considered “reliable” if the difference between the nominal and best estimate is considered acceptable with respect to prescribed dose limits or constraints, and anticipated levels of exposure. It follows that the notion of “reliability” is ultimately dependent on the exposure pathway in which the dose coefficient is applied. For example, a dose coefficient derived for a soluble form of a radionuclide will give higher doses from ingestion than the coefficient for an insoluble form. Consequently, it may be unduly conservative for protection purposes if such a dose coefficient is used to assess doses for an exposure pathway where the radionuclide is in an insoluble form. Reliability depends on two things: how close the nominal risk is to the best estimate of risk (relative to the dose limit), and how the dose coefficients are applied in practice.

Uncertainty analysis can be used to assess the first criterion by quantifying the range of the best estimate of the risk coefficient. It is clearly not possible, however, to use this

approach to assess the reliability of dose coefficients in every conceivable exposure scenario. Instead, a more general assessment of reliability can be made by limiting the described approach to dose coefficients that are used to assess doses from a number of the more relevant exposure pathways. With this in mind, the present report considers exposures for radioisotopes of a range of elements, including caesium, iodine, strontium, uranium, radium, plutonium, americium, and tritium. Exposure pathways are considered in a general sense – those that are likely to affect the general population. It should be noted that the internal doses that result from the ingestion or inhalation of these radionuclides are very small when compared with domestic exposures to radon progeny and other radiological hazards, particularly those resulting from diagnostic medical procedures. For example, average annual doses to the UK public resulting from ingestion and inhalation of natural uranium isotopes are less than 0.5 μSv , with similar levels being observed for other naturally occurring radionuclides (Watson et al 2005); this compares with an annual average dose of the order of 1300 μSv from domestic exposures to radon progeny. Nevertheless, the radionuclides considered in this report are of topical interest because of their association with industry, particularly the nuclear fuel cycle: with the exception of radium, the radionuclides are fission products or associated with nuclear fuel fabrication or reprocessing. Thus, these radionuclides are of more concern in areas where they may have accumulated in the environment as a result of industrial activity. Although it is beyond the scope of this report, the strategy described could be applied to assess the reliability of particular dose coefficients in a more localised exposure context. For example, and as discussed in section 3, public exposures to radium occur mainly through the ingestion of naturally occurring ^{226}Ra in food and water. However, higher levels of exposure through ingestion can potentially occur in areas where this radionuclide has been released into the environment as a result of localised industrial activity; for example, contamination arising from past radium luminising operations or from use of thorium in gas mantle production.

4.1 Assessing the reliability of dose coefficients using uncertainty analysis

A number of studies have been published that apply uncertainty analysis to quantify uncertainties on internal dose and risk resulting from the ingestion or inhalation of radionuclides. Only one study has attempted to quantify uncertainties on cancer risk coefficients resulting from internal exposure to environmental radionuclides (other than radon progeny). Pawel et al (2007) published an uncertainty analysis that compliments the risk coefficients published in FGR 13 (EPA 1999), for an “average” member of the US public. A simplified approach to that used in FGR 13 was used to calculate the uncertainty on the cancer mortality risk coefficient for a sample of radionuclides addressed in the original publication. Uncertainties on risk coefficients were taken from a joint NRC-CEC study that used expert elicitation techniques to predict 5, 50 and 95% values of the number of radiation induced cancer deaths expected over a lifetime in a hypothetical European/US population, assuming that each individual received an acute uniform whole body dose of 1 Gy of low LET radiation (NRC-CEC 1997, 1998). These were used in conjunction with distributions of tissue specific RBE values, DDREF, and

committed absorbed tissue doses (cumulative dose received up to 20 years after intake). The latter were typically calculated using a discrete set of biokinetic models that represented a plausible range of the biological behaviour of each radionuclide. Because of the large number of radionuclides that were considered, uncertainties on risk coefficients were grouped into categories, ranging from category A: the 90% range of values was within a factor of 15, to category E: the 90% range was greater than a factor of 150. Most radionuclides fell into category A or B (the 90% range of values was between a factor of 15 and 35 for category B). However, a limitation of this study is that it combines “plausible ranges” of biokinetic behaviour, with elements of probabilistic uncertainty analysis, and so provides indicative rather than probabilistic uncertainties on risk.

As in the present report, other studies have limited the application of parameter uncertainty analysis to calculate distributions of dose, often using the ICRP Publication 60 formalism (ICRP 1991) to calculate distributions of committed equivalent tissue doses and effective dose. Other studies, notably a recent study by NCRP (2010), have derived uncertainties on central values of effective dose based on expert elicitation techniques and a review of published studies. Interpretation of the distributions of effective dose calculated in such studies is often difficult: it is not clear, for instance, what is meant by terms such as “uncertainty on central values” or “population average” when referring to effective dose (Apostoaie and Miller 2004, NCRP 2010). Puncher and Harrison (2012a) reviewed studies that applied parameter uncertainty analysis to calculate distributions of equivalent tissue doses and effective dose, in order to derive nominal uncertainty factors for exposure to caesium, iodine and tritium. The present study extends the scope of these studies to:

- A. Consider the effects of uncertainty and variability in the derivation and propagation of parameter uncertainties into the distribution of dose for the purpose of inferring uncertainty on the location of the mean value of dose with respect to the dose coefficient; expressed as an “uncertainty factor”.
- B. Consider explicitly the combination of exposure pathway, which is particularly important for deriving distributions of values for material specific parameters, and the appropriate dose coefficient.
- C. Consider a wider range of radionuclides, particularly actinides.
- D. Consider exposures for different age groups.

The uncertainty factors for mean effective dose per unit intake, inferred from published studies by Puncher and Harrison (2012a) are compared with those obtained in the present study, in Table 4.1. Where a comparison is possible, the published studies are in good agreement with the analysis described in this report, and are consistent with UF values of around 2-3 for ingestion and 2-6 for inhalation, which result from uncertainties in the biokinetic model and model parameter values. The UF values derived in this study were the same for all age groups and are because the geometric (rather than absolute) range of the distributions of dose per unit intake was the same for each age group.

The table also compares the derived uncertainty factors with those inferred from the USEPA study (Pawel et al 2007). It should be noted that the factors inferred from that study reflect the geometric range of the risk coefficients and *not* the inferred location of the risk coefficient with respect to the nominal risk associated with the appropriate ICRP dose coefficient. The USEPA analysis calculated age and gender averaged risk coefficients but used an obsolete representation of the lung, and did not use a probabilistic approach when including biokinetic uncertainties. Given these limitations, it is not clear how to interpret the ranges of risk coefficients derived in the USEPA study; nevertheless the geometric ranges are reasonably consistent with the UF values obtained in this report when it is considered that the USEPA study includes uncertainties on the underlying cancer risk models relating averaged absorbed organ dose to cancer incidence and mortality, in addition to biokinetic uncertainty. The results of the USEPA study suggested that uncertainty in the risk model predictions, as opposed to uncertainty in the biokinetic models, accounted for a large proportion of the uncertainty in the cancer risk coefficients, although the proportion varied considerably between radionuclides (Pawel et al 2007). How indicative these results are for any particular population is difficult to determine because the uncertainties on risk estimates in that study pertain to a large hypothetical population (NCR/CEC 1997, 1998).

In this report and in the reviewed studies, uncertainties in the dosimetry model, which defines the spatial relationship between source and target tissues, were not included. This is due to the historical difficulty in producing probability distributions of absorbed fractions that reflect accurately the geometries of “true” reference humans or anatomical structures. This situation is likely to change with the recent emergence of more anatomically realistic phantoms. A useful review of uncertainties in current dosimetry models is provided by NCRP (2010).

Table 4.1 Comparison of uncertainty factors for dose coefficients used to protect adult members of the public.^a

Radionuclide	Source ^b	Route of intake	Uncertainty Factor		
			This study	Others	USEPA
¹³⁷ Cs	3	Ingestion	<3	2	<5
¹³⁷ Cs	-	Inhalation	3	-	6-7
⁹⁰ Sr	3	Ingestion	3	3	<5
⁹⁰ Sr	-	Inhalation	3	-	5-6
²³⁸ U	-	Ingestion	3	-	5-6
²³⁸ U	-	Inhalation	2	-	7-9
²²⁶ Ra	-	Ingestion	3	-	7-9
²²⁶ Ra	-	Inhalation	6	-	5-6
²³⁹ Pu	8	Ingestion	3	3	7-9
²³⁹ Pu	-	Inhalation	<3	-	5-6
²⁴¹ Am	8	Ingestion	3	3	7-9
²⁴¹ Am	-	Inhalation	<3	-	5-6
³ H (HTO)	6, 7	Inhalation/ingestion	2	2	<5
³ H (OBT)	6, 7	Ingestion	2	<3	5-6
¹³¹ I	2, 3, 4, 5	Ingestion	2	2	6-7
¹³¹ I	1, 2	Inhalation	2	<3	7-9
¹²⁹ I	2	Ingestion	<4	6	7-9
¹²⁹ I	-	Inhalation	<4	-	7-9

^aUncertainty factors inferred from the analyses conducted in this report ("This study") are compared with those inferred in the review of Puncher and Harrison (2012a) (column "Others"). These are compared with uncertainty factors inferred from the USEPA study of cancer mortality risk coefficients (column "USEPA"); note that the cancer risk coefficients are for an "age and gender averaged" individual based on a hypothetical US population, and the given values refer to the range of risk coefficients.

^b¹Harvey and Hamby (2006); ²Fritsch (2007); ³Apostoaie and Miller (2004); ⁴Dunning and Schwarz (1981); ⁵Hamby and Benke (1999); ⁶Harrison et al (2002); ⁷Melintescu et al (2007); ⁸Puncher and Harrison (2012b)

5 SUMMARY AND CONCLUSIONS

ICRP is clear on the intended use of equivalent and effective dose as reference quantities, without uncertainty, for use in internal radiation protection. However, ICRP and others also recognise that there are uncertainties in the process of estimating dose and risk that affect the derivation and application of these quantities. This report addresses this issue as follows:

1. The issue of relevance for regulators and other stakeholders is not the magnitude of the "uncertainty" on dose estimates, but how "reliable" dose coefficients are for protection purposes, as a protection device. It is argued that a dose coefficient, as applied to a defined exposure pathway, is considered reliable if it ensures exposures comply with dose limits and constraints.

2. The best estimate of risk and its uncertainty, for a given internal exposure pathway, is a pre-requisite to making an informed judgement on the reliability of a particular dose coefficient in the context of a specified exposure pathway. More precisely, it is the uncertainty associated with the best estimate of risk, with respect to the nominal risk associated with the dose coefficient, which is sought. In cases where it is difficult to quantify this range directly, it may be more conveniently represented as an inferred “uncertainty factor” or UF, which indicates a subjectively derived 95% probability interval for this range.
3. A general assessment of the reliability of dose coefficients can be made by assessing the reliability of dose coefficients that are applied to the more significant exposure pathways.
4. With the idea of general assessment in mind, parameter uncertainty analysis is performed for some of the most significant radionuclides for public and environmental protection purposes. The analysis considers uncertainties in the biokinetic models and model parameter values, and propagates these distributions into the calculation of effective dose per unit intake. The distributions are used to infer the value of uncertainty factors for the appropriate dose coefficients, with respect to biokinetic uncertainties
5. The analyses indicate UF values of around 2-3 for ingestion, and 2-6 for inhalation. The UF values are the same for the three age groups considered: 1 year olds, 10 year olds and adults. The UF values are consistent with those inferred from other studies.
6. Although a general assessment of reliability of the protection quantities is beyond the scope of this report, the derived UF values for the radionuclides considered here seem acceptable when considered alongside the likely levels of exposure that is expected from them (the sub micro-sievert to micro-sievert range) and the dose limit for planned exposures for members of the public: 1000 micro-sieverts; and viewed in context with all radiological hazards to the general public: an estimated average annual dose of 2700 micro-sieverts per year in the UK, incurred mostly from radon and other natural sources of radiation, and medical exposures.
7. The exposure pathways considered in the analysis represent general environmental exposure to members of the UK public. However, the derived uncertainties, at least for non-material specific parameters, can be directly applied to consider other exposure scenarios as required.
8. The described strategy can be readily extended to include uncertainties in the estimation of cancer risk, and other parameters, for a more comprehensive assessment of uncertainty on risk and assessment of reliability of dose coefficients.

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7 REFERENCES

- Agency for Toxic Substances and Disease Registry (ATSDR 2011). Draft Toxicological Profile for Uranium; available from www.atsdr.cdc.gov/toxprofiles/tp150.pdf (Accessed 20th February 2013).
- Alexandrou, A. Uncertainties on doses from ingested and inhaled uranium. MSc dissertation, University of Surrey (2010).
- Apostoaiei, A. I. Absorption of strontium from the gastro-intestinal tract into plasma in healthy human adults. *Health Phys.* 83:56-65 (2002).
- Apostoaiei, A.I., Miller, L.F. Uncertainties in dose coefficients from ingestion of ^{131}I , ^{137}Cs and ^{90}Sr . *Health Phys.* 86, 460-482 (2004).
- Bailey, M.R., et al. Updating the ICRP Human Respiratory Tract Model. *Radiat. Prot. Dosim.* 127. 31-34 (2007).
- Bailey, M.R., Puncher, M. Uncertainty analysis of the ICRP Human Respiratory Tract Model applied to the interpretation of bioassay data for depleted uranium. Report HPA-RPD-023, Health Protection Agency, Chilton (UK), (2007).
- Bassett, S.H., Frankel, A., Cedars, N., et al. The excretion of hexavalent uranium following intravenous administration: II. Studies on human subjects. Rochester, New York, UR-37 (1948).
- Birchall, A., James, A.C. Uncertainty analysis of the effective dose per unit exposure from radon progeny and implications for ICRP risk-weighting factors. *Radiat. Prot. Dosim.* 53(1-4), 133-140 (1994).
- Birchall, A., Puncher, M., Marsh, J.W., et al. IMBA Professional Plus: a flexible approach to internal dosimetry. *Radiat. Prot. Dosim.* 125; 194-197 (2007).
- Birchall, A., Puncher, M., Harrison, J. D., Riddell, A., Bailey, M. R., Khokryakov, V. and Romanov, S.. Plutonium worker dosimetry. *Radiat. Env. Biophys.* 49(2), 203-212 (2010).
- Blanchardon, E., Leggett, R.W., Eckerman, K.F. Some elements for a revision of the americium reference biokinetic model. *Radiat. Prot. Dosim* 127(1-4), 131-135 (2007).
- Carlin, B.P., Louis, T.A. Bayes and empirical bayes methods for data analysis. Chapman & Hall/CRC texts in statistical science (Chatfield and Zidek eds.) ISBN 58488-170-4; Chapman and Hall, NY (2000).
- CERRIE. Report of the Committee Examining Radiation Risks of Internal Emitters. Avail. from Health Protection Agency, CRCE, Chilton, UK. ISBN 0-85951-545-1. (2004).
- Clewell, H. J., et al. Quantitative Interpretation of Human Biomonitoring Data. *Tox. Appl. Pharmacol.* 231, 122-133 (2008).
- Collins, C. D., Gravett, A. E., Bell, J. N. B. The deposition and translocation of methyl iodide by crops. *Health Phys.* 87(5):51-517 (2004).

- Cuddihy, R. G., Finch, G. L., Newton, G. J., Hahn, F. F., Mewhinney, J. A., Rothenberg, S. J. and Powers, D. A. Characteristics of radioactive particles released from the Chernobyl nuclear reactor. *Environ. Sci. Technol.* 23, 89–95 (1989).
- Davesne E, Paquet F, Ansoborlo E, Blanchardon E. Absorption of plutonium compounds in the respiratory tract. *J Radiol Prot*; 30:5–21 (2010).
- Dorrian, M-D. Particle size distributions of radioactive aerosols in the environment. *Radiat. Prot. Dosim.* 69(2) 117-132 (1997).
- Dunning, D.E., Schwartz, G. Variability of human thyroid characteristics and estimates of doses for ingested I-131. *Health. Phys.* 40, 661-75 (1981).
- Dua, S. K., Maniyan, C. G. and Kotrappa, P. Inhalation exposures during operations in spent fuel bays. *Radiat. Prot. Dosim.* 19, 165–172 (1987).
- Eastell, R., Delmas, P.D., Hodgson, S.F., Eriksen, E.F., Mann, K.G., Riggs, B.L. Bone formation rate in older normal women: concurrent assessment with bone histomorphometry, calcium kinetics, and biochemical markers. *J. Clin. Endocrinol. Metab.* 67(4), 741-8, 1988.
- Environment Agency (EA). Radioactivity in food and the environment (RIFE), report 2010. Available from: <http://www.environment-agency.gov.uk/homeandleisure/> (accessed April 2012).
- Environmental Protection Agency (EPA). Cancer risk coefficients for environmental exposures to radionuclides; Federal Guidance Report 13. US Environmental Protection Agency, Washington DC. (1999).
- Fatayerji, D., Eastell, R. Age-related changes in bone turnover in men. *J. Bone Miner. Res.* 14,1203-1210 (1999).
- Fell, T. P. The computation of ICRP dose coefficients for intakes of radionuclides with PLEIADES: biokinetic aspects. *Radiat. Prot. Dosim.* 127(1-4); 200-222 (2007).
- Fritsch, P. Uncertainties in committed equivalent doses to the thyroid after ingestion or inhalation of different chemical forms 125-129-131I. *Radiat. Prot. Dosim* 127(1-4); 548-552 (2007).
- Galeriu, D., Melintescu, A. Retention of tritium in reference persons: a metabolic model. Derivation of parameters and application of the model to the general public and to workers. *J. Radiol. Prot.* 30, 445-468 (2010).
- Gregoratto, D., Bailey, M.R., Marsh, J.W. Particle retention in the alveolar–interstitial region: proposed changes to the Human Respiratory Tract Model. *J. Radiol. Prot.* 30, 491-512 (2010).
- Griffis, L. C., Snipes, M. B. Clearance of coal combustion fly ash from rat lungs after acute inhalation exposure. *Inhalation Toxicology Research Institute Annual Report 1980-1981*, Albuquerque, NM 87185, USA; prepared for the US Department of Energy. (1981).
- Hamby, D.N., Benke, R.R. Uncertainties of the I-131 ingestion dose conversion factor. *Radiat. Prot. Dosim.* 82(4) 245–256 (1999)
- Harrison, J.D., Leggett, R.W., Noßke, D., Paquet, F., Phipps, A.W., Taylor D.M., Metivier, H. Reliability of the ICRP's dose coefficients for members of the public, II. Uncertainties in the absorption of ingested radionuclides and the effect on dose estimates. *Radiat. Prot. Dosim* 95, 295-308 (2001).
- Harrison, J.D., Khursheed, A., Lambert, B.E. Uncertainties in dose coefficients for intakes of tritiated water and organically bound forms of tritium by members of the public. *Radiat. Prot. Dosim.* 98(3): 299-311 (2002).
- Harrison, J.D, Fell, T. P., Pellow, P. G. D., et al. Uncertainty analysis of the ICRP systemic model for uranium as applied to the interpretation of bioassay data for depleted uranium. Report HPA-RPD-024, Health Protection Agency, Chilton (UK); (2007).
- Harrison, J.D., Day, P. Radiation doses and risks from internal emitters. *J. Radiol. Prot.* 28(2), 137-159 (2008).
- Harvey, R.P., Hamby, D.M., Palmer, T.S. Uncertainty of the thyroid dose conversion factor for inhalation intakes of 131I and its parametric uncertainty. *Radiat. Prot. Dosim.* 118(3): 296-306 (2006).

- Hill, R.L., Johnson, J.R. Metabolism and dosimetry of tritium. *Health Phys.* 65, 628-647 (1993).
- Hunt, G.J., Leonard, D.R.P., Lovett, M.B. Transfer of environmental plutonium and americium across the human gut: a second study. *Tot Sci Env* 90, 273-282 (1990).
- Hunt, G.J., Leonard, D.R.P., Lovett, M.B.. Transfer of environmental plutonium and americium across the human gut. *Tot. Sci. Env.* 53, 89-109 (1986).
- Hunt, G.J. Transfer across the human gut of environmental plutonium, americium, cobalt, caesium and technetium: studies with cockles (*Cerastoderma edule*) from the Irish Sea. *J Radiol Prot* 18(2), 101-109 (1998).
- ICRP. Limits for intakes of radionuclides by workers. ICRP Publication 30; Ann. ICRP 2(3/4) (Oxford: Pergamon Press) (1979).
- ICRP. Age-dependent doses to members of the public from intake of radionuclides: part 1. ICRP Publication 56; Ann. ICRP 20(2) (Oxford: Pergamon Press) (1989).
- ICRP. 1990 Recommendations of the International Commission on Radiological Protection. ICRP Publication 60; Ann. ICRP 21(1-3) (Oxford: Pergamon Press) (1991).
- ICRP. Age-dependent doses to members of the public from intake of radionuclides: Part 2 Ingestion dose coefficients. ICRP Publication 67; Ann ICRP 23(3/4). Oxford:Pergamon press (1993).
- ICRP. Dose coefficients for intake of radionuclides by workers. ICRP Publication 68; Ann. ICRP 24(4) (Oxford: Elsevier Science Ltd (1994a).
- ICRP. Human respiratory tract model for radiological protection. ICRP Publication 66; Ann ICRP 24(1-3). (Oxford: Pergamon press; (1994b).
- ICRP. Age-dependent doses to members of the public from intake of radionuclides: Part 4 Inhalation dose coefficients. ICRP Publication 71; Ann ICRP 25(3/4). Oxford:Pergamon press (1995a).
- ICRP. Age-dependent doses to members of the public from intake of radionuclides: Part 3 Ingestion dose coefficients. ICRP Publication 69; Ann ICRP 25(1). Oxford:Pergamon press (1995b).
- ICRP. Age-dependent doses to members of the public from intake of radionuclides: Part 5 Compilation of ingestion and Inhalation dose coefficients. ICRP Publication 72; Ann ICRP 26(1). Oxford:Pergamon press (1996).
- ICRP. Basic Anatomical and Physiological Data for Use in Radiological Protection Reference Values. ICRP Publication 89; Ann. ICRP 32(3-4) (Oxford: Elsevier Science Ltd.) (2002).
- ICRP. Assessing dose of the representative person for the purpose of radiation protection of the public and the optimisation of radiological protection: broadening the process. ICRP Publication 101; Ann. ICRP 36(3) (Oxford: Elsevier Science Ltd.) (2006a).
- ICRP. Human alimentary tract model for radiological protection. ICRP Publication 100; Ann. ICRP 36(1-2) (Oxford: Pergamon Press) (2006b).
- ICRP. The 2007 recommendations of the international commission on radiological protection. ICRP Publication 103; Ann. ICRP 37(2-4) (Oxford: Elsevier Science Ltd.) (2007).
- ICRP. Adult reference computational phantoms. ICRP Publication 110; Ann. ICRP 39(2) (Oxford: Pergamon Press) (2009).
- International Organization for Standardization. Guide to the expression of uncertainty in measurement (GUM) ISO/IEC Guide 98 (1993).
- Kalkwarf, D.R., Jackson, P.O., Hardin, J.M. Lung-clearance classification of radionuclides in coal fly ash. *Health Phys.*, 47; 37-45 (1984).
- Killough, G.G., Eckerman, K.F. Age and sex-specific estimation of dose to a normal thyroid from clinical administration of ¹³¹I. Oak Ridge, TN: ORNL/TM-9800, NUREG/CR-3955 (1986).
- Khokhryakov VF, Suslova KG, Vostrotin VV, Romanov RA, Eckerman KF, Krahenbuhl MP, et al. Adaptation of the ICRP Publication 66 respiratory tract model to data on plutonium biokinetics for Mayak workers. *Health Phys* 2005; 88:125-32.
- Khursheed, A., Fell, T.P. Sensitivity analysis for the ICRP Publication 67 biokinetic model for plutonium. *Radiat. Prot. Dosim.* 74, 63-73 (1997).

- Kutkov, V. A. and Komaritskaya, O. I. In vitro solubility of Chernobyl nuclear fuel aerosol with respect to collective behaviour of its radionuclides. In : Proc. IRPA 9, Vienna, 2, 445-447 (1986).
- Kutkov, V. A. Application of human respiratory tract models for reconstruction of the size of aerosol particles through the investigation of radionuclides behaviour in the human body. Radiat. Prot. Dosim. 79, 265–268 (1998).
- Kutkov, V. A. Results of in vivo monitoring of the witnesses of the Chernobyl accident. Radiat. Prot. Dosim. 89, 193-197 (2000).
- Leggett, R.W. A model of the retention, translocation and excretion of systemic Pu. Health Phys. 49(6), 1115-1137 (1985).
- Leggett, R. W. Predicting the retention of cesium in individuals. Health Phys 50, 747-759 (1986).
- Leggett, R.W. A generic age-specific biokinetic model for calcium-like elements. Radiat. Prot. Dosim 41(3); 183-198 (1992a).
- Leggett, R.W. A retention-excretion model for americium in humans. Health Phys. 62(4), 288-310 (1992b).
- Leggett, R.W. Basis for the ICRP's age-specific biokinetic model for uranium. Health Phys. 67; 589-610 (1994).
- Leggett, R.W. Reliability of the ICRP's dose coefficients for members of the public. 1. Sources of uncertainty in the biokinetic models. Radiat. Prot. Dosim. 95(3), 199-213 (2001).
- Leggett, R. W., et al. A physiologically based model for cesium in the human body. Sci. Tot. Environ 317, 235-255 (2003).
- Leggett, R. W., Eckerman, K. F., Khokhryakov, V. F., Suslova, K. G., Krahenbuhl, M. P. and Miller, S. C. Mayak worker study: an improved biokinetic model for reconstructing doses from internally deposited plutonium. Rad. Res. 164, 111-122, (2005).
- Leggett, R. W. A physiological systems model for iodine for use in radiation protection. Radiat. Res. 174, 496-516 (2010).
- Leggett, R. W., Harrison, J. D. Fractional absorption of ingested uranium in humans. Health Phys 68, 1-15 (1995).
- Li, W. B., Hollriegel, V., Roth, I., Oeh, U. Human biokinetics of strontium. Part I: Intestinal absorption rate and its impact on the dose coefficient of ^{90}Sr after ingestion. Radiat. Environ. Biophys. 45: 115–124 (2006).
- Mckay, M.D., Beckman, R.J., Conover, W.J. A comparison of three methods for selecting values of input variables in the analysis of output from a computer code. Technometrics 21, 239-245 (1979).
- Melintescu, A., Galeriu, D., Takeda, H. Reassessment of tritium dose coefficients for the general public. Radiat. Prot. Dosim 127(1-4) 153-157 (2007).
- Miller G. Variability and uncertainty of biokinetic model parameters: the discrete empirical bayes approximation. Radiat. Prot. Dosim. 131(3); 394-398 (2008).
- National Council on Radiation Protection and Measurements (NCRP). Tritium in the environment. NCRP Report 62. Washington, D.C. (1979).
- Nair, S., Pradhan, A.S., Jaiswal, D. D., Singh, I. S., Garg, S. P. Comparison of observed skeleton retention of strontium in average indian adult with the value predicted by the ICRP biokinetic model Radiat. Prot. Dosim. 118(4): 475-478 (2006)
- NCRP. A guide for uncertainty analysis in dose and risk assessments related to environmental contamination. NCRP Commentary 14. NCRP Bethesda, MD 20814-3095. (1996).
- NCRP. Deposition, retention and dosimetry of inhaled radioactive substances. Baltimore, MD: National Council on Radiation Protection and Measurements; NCRP Report No. 125; (1997).
- NCRP. Evaluating the reliability of biokinetic and dosimetric models and parameters used to assess individual doses for risk assessment purposes. NCRP Commentary 15. NCRP Bethesda, MD 20814-3095. (1998).

- NCRP. Uncertainties in internal radiation dose assessment. NCRP Report 164. NCRP Bethesda, MD 20814-3095. (2010).
- National Regulatory Commission – Commission of European Communities. Probabilistic accident consequence uncertainty analysis: late health effects uncertainty assessment. NUREG/CR-6555; EUR 16774; SAND97-2322. US Nuclear Regulatory Commission, Washington DC; Office for publications of the European Communities, Luxembourg (1997).
- National Regulatory Commission – Commission of European Communities. Probabilistic accident consequence uncertainty analysis: uncertainty assessment for internal dosimetry. NUREG/CR-6571; EUR 16773; SAND98-0119. US Nuclear Regulatory Commission, Washington DC; Office for publications of the European Communities, Luxembourg (1998).
- NRC. Health Risks from Exposure to Low Levels of Ionizing Radiation. Biological Effects of Ionizing Radiation (BEIR) VII Phase 2. National Research Council of the National Academies. The National Academies Press, Washington DC. (2006).
- Nguyen, T.V., Meier, C., Center, J.R., Eisman, J.A., Seibe, I.M.J. Bone turnover in elderly men: relationships to change in bone mineral density. *BMC Musculoskeletal Disorders* doi:10.1186/1471-2474-8-13 (2007).
- Niinimaa, V., Cole, P., Mintz, S., Shephard, R.J. The switching point from nasal to oronasal breathing. *Respir Physiol* 42, 61-71, (1980).
- Niinimaa, V., Cole, P., Mintz, S., Shephard, R.J. Oro-nasal distribution of respiratory airflow. *Respir. Physiol* 43: 69-75, (1981).
- Pawel, D., J., Leggett, R. W., Eckerman, K.F., Nelson, C. B. Uncertainties in cancer risk coefficients for environmental exposure to radionuclides: an uncertainty analysis for risk coefficients in federal guidance report no. 13. Oak Ridge National Laboratory. ORNL/TM-2006/583 (2007).
- Phipps, A. W., Fell, T.P., et al. Dose coefficients calculated using the new ICRP model for the human alimentary tract. *Radiat. Prot. Dosim.* 127(1-4), 79-85 (2007).
- Polig, E., Lloyd, R.D., Bruenger, F., W., Miller, S.C. Biokinetic model of radium in humans and beagles. *Health Phys.* 86(1); 42-55 (2004).
- Puncher, M., Birchall, A. Estimating uncertainty on internal dose assessments. *Radiat. Prot. Dosim.* 127, 544-547 (2007).
- Puncher, M., Bailey, M.R. and Harrison, J.D. Uncertainty analysis of doses from inhalation of depleted uranium. *Health Phys.* 95(3), 300 –309 (2008).
- Puncher, M., Birchall, A., Bull, R.K. Uncertainties on lung doses from inhaled plutonium. *Radiat. Res.* 176, 494-507 (2011).
- Puncher, M., Harrison, J. D. Assessing the reliability of dose coefficients for inhaled and ingested radionuclides. *J. Radiol. Prot.* 32, 223–241 (2012a).
- Puncher, M., Harrison, J. D. Uncertainty analysis of doses from ingestion of plutonium and americium. *Radiat. Prot. Dosim.* 148(3), 284-296 (2012b).
- Reddy, M., B., Yang, R. S., et al. Physiologically based pharmacokinetic modelling: science and applications. Wiley-Interscience (2006).
- Richardson, R. B., Dunford, D. W. A biochemical –based model for the dosimetry of dietary organically bound tritium – part 1: physiological criteria. *Health Phys.* 85(5), 523-538 (2003a).
- Richardson, R. B., Dunford, D. W. A biochemical –based model for the dosimetry of dietary organically bound tritium – part 2: dosimetric evaluation. *Health Phys.* 85(5), 539-552 (2003b).
- Riggs, D. S. Quantitative aspects of iodine metabolism in man. *Pharmacol. Rev* 4, 284-370 (1952).
- Saito, M. A modified three-compartment model for tritium metabolism in man. *Rad. Prot. Dosim.* 42, 1017-1024 (1992).
- Schlenker, R.A., Keane, A.T., Holtzman, R.B. The retention of Ra-226 in human soft tissue and bone; implications for the ICRP 20 alkaline earth model. *Health Phys.*, 42; 671-693 (1982).

- Shagina, N. B., Tolstykh, E., Degteva, M. O. Improvements in the biokinetic model for strontium with allowance for age and gender differences in bone mineral metabolism. *Radiat. Prot. Dosim.* 105(1-4): 619-622 (2003).
- Stradling, G. N., Pellow, P. G., Hodgson, A., Fell, T. P., Phipps, A., Pearce, M., Rance, E., Ellender, M., Taskaeva, M., Penev, I. and Guentchev, T. Dose coefficient and assessment of intake of a radionuclide bearing dust formed at the Kozloduy Nuclear Power Plant, NRPB-M679. Chilton, National Radiological Protection Board. (1996).
- Stradling, G. N., Pellow, P. G., Hodgson, A., Fell, T. P., Phipps, A., Pearce, M., Taskaeva, M., Penev, I. and Guentchev, T. Assessment of intake of a complex radionuclide bearing dust formed at a nuclear power plant. *J. Radioanal. Nucl. Chem.* 226 1-2, 7-14 (1997).
- Suslova, K.G., et al. Extrapulmonary organ distribution of plutonium in healthy workers exposed by chronic inhalation at the Mayak Production Association. *Health Phys.* 82, 432-444 (2002).
- Smith, K. R., Crockett, G. M., Oatway, W. W., et al. Radiological impact on the UK population of industries which use or produce materials containing enhanced levels of naturally occurring radionuclides: Part I: Coal-fired electricity generation. Report of the UK National Radiological Protection Board (NRPB), NRPB-R327; NRPB, Chilton, UK; ISBN 0 85951 462 5, (2001).
- Smith, J.R.H., Bailey, M. R., Etherington, G., Shutt, A.L., Youngman, M.J. Further study of the effect of particle size on slow particle clearance from the bronchial tree. *Radiat. Prot. Dosimetry* 127(1-4): 35-39 (2007).
- Smith, J.R.H., Bailey, M. R., Etherington, G., Shutt, A.L., Youngman, M. An experimental study of clearance of inhaled particles from the human nose. *Exp. Lung Res.* 37(2), 109-129 (2011)
- Spiegelhalter, D.J., Abrams, K.R., Myles, J.P. Bayesian approaches to clinical trials and health care evaluation. (Statistics in Practice; eds Senn S, Barnett V); Wiley and Sons, Chichester (UK) (2004).
- Stanbury, S. B., et al. The adaptation of man to iodine deficiency. In: endemic goitre. Cambridge, MA: Harvard University Press, (1954).
- Stayner, L., Vrijheid, M., Cardis, E., Stram, D. O., Deltour, I., Gilbert, S. J. and Howe, G. A monte carlo maximum likelihood method for estimating uncertainty arising from shared errors in exposures in epidemiological studies of nuclear workers. *Radiat. Res.* 168, 757–763 (2007).
- Struxness, E. G., Luessenhop, A. J., Bernard, S. R., Gallimore, J.C. The distribution and excretion of hexavalent uranium in man. IN Proceedings of the international conference on the peaceful uses of atomic energy. 8th Aug 1995 to 20th Aug 1955. New York, United Nations, 186-196 (1955).
- Talbot, R. J., D., et al. Sex-related differences in the human metabolism of Pu. *Radiat. Prot. Dosim.* 71, 107–121 (1997).
- Taylor, D. M. A biokinetic model for predicting the retention of ³H in the human body after intakes of tritiated water. *Radiat. Prot. Dosim.* 105(1-4), 225-228 (2003).
- Taylor, D. M. Biokinetic models for the behaviour of carbon-14 from labelled compounds in the human body: can a single generic model be justified? *Radiat. Prot. Dosim.* 108(3), 187–202 (2004).
- Terepka, A.R., Toribara, T.Y., Neuman, W.F. Abstract 22: Skeletal retention of uranium in man IN 46th meeting of the endocrine society (1964).
- United Nations Scientific Committee on the Effects of Atomic Radiation 2006 report: Effects of ionising radiation – volume 1. United Nations, New York (2006).
- Vanderpump, M.P.J., et al. Iodine status of UK schoolgirls: a cross-sectional survey. *Lancet.* 377, 2007-12 (2011).
- Watson, S. J., Jones, A. L., Oatway, W. B., Hughes, J. S. Ionising radiation exposure of the UK population: 2005 review. HPA-RPD report 001, Health Protection Agency, Chilton, UK (ISBN 0 85951 558 3) (2005).
- Wayne, E. J., et al. Clinical aspects of iodine metabolism. Oxford, England: Blackwell Scientific Publications, (1964).
- Whillans, D. W. Structure of a physiologically based biokinetic model for use in ¹⁴C and organically bound tritium dosimetry. *Radiat. Prot. Dosim* 105(1-4), 189-191 (2003).

World Health Organization (WHO). Iodine deficiency in Europe: a continuing public health concern (Geneva: World Health Organization) (2007).

Zvonova, I. A. Dietary intake of stable I and some aspects of radioiodine dosimetry. *Health Phys.* 57, 471-475 (1989).

APPENDIX A

Calculating a population specific risk coefficient

Calculating the cancer risk coefficient for a given population requires that the risk coefficient be calculated for each $g=1$ to G sub-group, defined by age and gender, in the exposed population. The steps are as follows:

1. Average absorbed tissue doses (in Gy) are calculated using appropriate biokinetic and dosimetric models, for a unit intake by ingestion or inhalation. Because internal doses to tissues from inhaled or ingested radionuclides can be received over a protracted time period (a time interval defined by the physical half life or biological residence time of the radionuclide) and the cancer risk to tissues varies with age at exposure, average absorbed tissues doses are calculated for each year following the intake, rather than cumulated over a long time period, as is performed during the calculation of effective dose (see step 3).
2. Absorbed tissue doses are weighted according to the effectiveness of each radiation type in inducing cancer in irradiated tissues, relative to low LET radiation.
3. For each population sub-group, g , the lifetime risk of cancer incidence, R_t , incurred by each tissue, t , that results from an acute unit intake is calculated. This is achieved by combining the weighted dose received in each year following the intake (step 2), with best estimates of the risk of lifetime cancer incidence per unit absorbed dose (risk per gray) received in each year, and the survival function of population group, g . The best estimates of lifetime cancer incidence per unit absorbed dose by age at exposure are extrapolated from studies of other populations exposed to ionising radiation. These include principally the life span study (LSS) of the Japanese atomic bomb survivors, (NRC 2006; UNSCEAR 2006; ICRP 2007). If the ultimate purpose of calculating a best estimate of risk is to assess the “reliability” of a dose coefficient (described in the next section), then the best estimate of cancer incidence for each tissue should be further adjusted for lethality, quality of life lost and heritable effects using the appropriate ICRP formalism (ICRP 2007).
4. The tissue specific risk coefficients are summed over all tissues, T , to obtain the total lifetime excess absolute cancer risk, R_g , per unit intake for population group, g :

$$R_g = \sum_{t=1}^T R_t \quad (\text{A.1})$$

The population specific cancer risk coefficient, R , is obtained by taking the mean of the risk coefficients calculated for each of the G population groups as follows:

$$R = \sum_{g=1}^G f_g R_g \quad (\text{A.2})$$

Where f_g is the fraction of individuals in population group, g .

The above summarises the stages in the calculation of a risk coefficient. Full details describing the derivation of cancer risk coefficients for radionuclide exposures are provided in chapter 7 of Federal Guidance Report (FGR) 13 (EPA 1999). In that report, the risk coefficients were averaged over a large population group whose composition, survival functions and cancer mortality rates were modelled on those of a representative US population. The calculations accounted for age and gender specific variations in the rate of exposure, including the consumption of radionuclides in food and water, and the pattern of intake (acute versus chronic). Because they are average quantities, the risk coefficients were intended for assessments of risk to large actual or hypothetical populations, rather than to specific individuals (EPA 1999).

APPENDIX B

Biokinetic models

B1 INTAKE TRACT MODELS

These are mathematical models that describe the deposition in the lungs or transit through the alimentary tract, of inhaled or ingested radionuclides, and their clearance to blood.

B1.1 The human respiratory tract model

The Human Respiratory Tract Model (HRTM) published by ICRP describes the deposition, clearance, and dosimetry of inhaled material in the lungs (ICRP 1994b) and has been widely used to calculate dose coefficients for workers and members of the public for intakes by inhalation (ICRP 1994a; 1995a). The model provides a suitable compromise between physiological realism and ease of practical application so that, in addition to its application in radiation protection, the model has been applied to calculate lung doses in epidemiology studies of nuclear workers exposed to plutonium and other actinides. However, the emergence of new experimental data, as well as a review of existing data, has prompted a proposed updating of the HRTM (Bailey et al 2007). These changes apply to the compartments representing particle transport clearance from the bronchial (BB), bronchiolar (bb), and alveolar-interstitial (AI) regions of the lung (Bailey et al 2007); and deposition and clearance from the extra-thoracic (ET) region. ICRP is considering this revised structure in its forthcoming publication on the Occupational Intakes of Radionuclides (OIR) which will provide revised dose coefficients for workers based on the updated ICRP Publication 103 (ICRP 2007) formalism for effective dose (Bailey et al 2007). The revised particle transport clearance model is given in Figure B.1; descriptions of the proposed structural changes are provided elsewhere (Bailey et al 2007; Gregoratto et al 2010; Smith et al 2007, 2011). In the revised model it is assumed that, of the material deposited in the ET region, 65% is deposited in ET1 and 35% in the ET2 region. In this analysis, the revised HRTM was assumed to be a more accurate representation of the deposition and clearance of inhaled material in the lungs than the original HRTM (ICRP 1994b), and was used in preference to it.

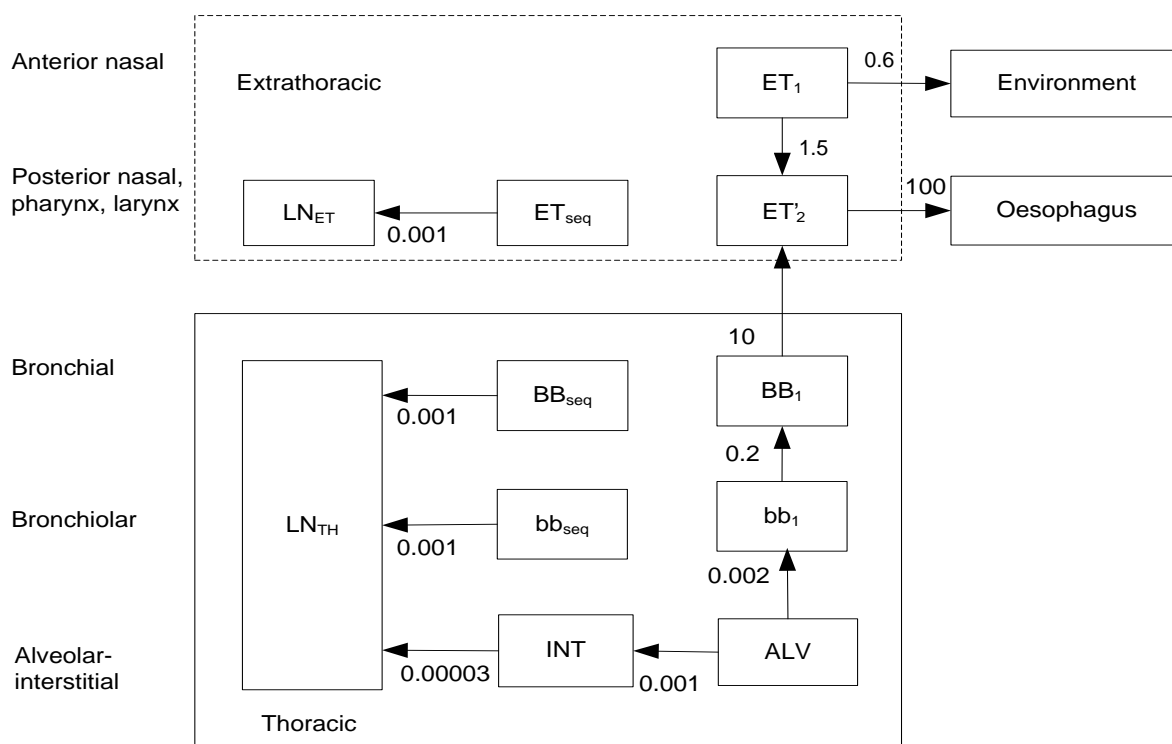


Figure B.1. Revised compartment model representing time-dependent particle transport from each respiratory tract region: the thoracic airways (lungs) consist of the alveoli (ALV), pulmonary interstitial tissue (INT), the bronchiolar airways (bb) and bronchial airways (BB); the extra-thoracic airways (ET) consist of the anterior nasal passage (ET1) and the posterior nasal passages (ET2). Rates shown alongside arrows are reference values in units of d^{-1} . It is assumed that 0.2% of material deposited in regions ET2, BB and bb is retained in the airway wall (ET_{seq}, BB_{seq} and bb_{seq} respectively).

B1.2 The alimentary tract model

ICRP has recently published a new biokinetic model describing the biokinetics and dosimetry of the alimentary tract (ICRP 2006). The new model supersedes the simpler four compartment model described in the earlier ICRP Publication 30 (ICRP 1979). The new model includes some additional compartments, principally the oral cavity and oesophagus, which were omitted in the original. It also includes recycling pathways between the alimentary tract contents and walled structures. However, for most radionuclides, including uranium, there is no recycling and the model reduces to a much simpler form.

In the ICRP Publication 30 model, the fraction of ingested material absorbed to blood from the alimentary tract is determined by the value of the f_1 parameter (ICRP 1979), which represents the fraction of material absorbed from the small intestine. In the new model (ICRP 2006), provision is made to account for absorption in other parts of the alimentary tract including the oral cavity, stomach and any segment of the colon, in addition to the small intestine. In the new model, the fraction of ingested material absorbed to blood, f_A , represents the sum of the values of the fractions assigned to each of these regions. However, in the majority of cases, it is anticipated that

insufficient information will be available to assign a fraction to regions other than the small intestine. Under these circumstances the f_A and f_1 parameter are equivalent.

The revised model also includes an updated dosimetry model, which describes the deposition of energy in target regions of the alimentary tract resulting from radioactive disintegrations that occur elsewhere in the body (described by the absorbed fractions, AF). However, the revised AFs for the model are not yet available. A preliminary analysis by Phipps et al (2007), using prototype AFs, indicates that for many radionuclides the revised model leads to doses that are similar to, or lower than, those predicted by the ICRP Publication 30 model. For the purpose of this project, it was assumed that the ICRP Publication 30 model was an adequate representation of the alimentary tract.

B2 SYSTEMIC MODELS

The systemic distribution of radionuclides is represented by first order compartment models. Although not strictly physiologically based pharmacokinetic (PBPK) models, many of the models, particularly those describing the kinetics of metals, have pathways associated with clearly defined physiological processes such as bone remodelling and hepatocyte turnover. An example is provided in Figure B.2, for the model describing the kinetics of strontium and radium.

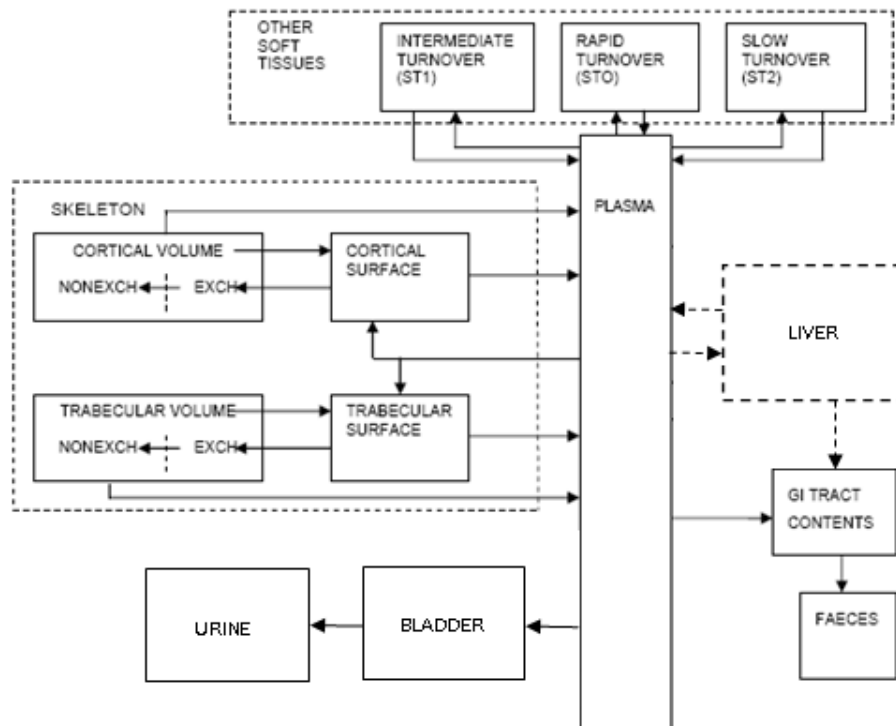


Figure B.2. The ICRP Publication 67 systemic model for strontium and radium. The liver compartment is not included in the model for strontium.

B2.1 Strontium and Radium

The ICRP systemic models for strontium and radium are based on a generic structure derived for the alkaline-earth metals, and physiological analogues lead and uranium (ICRP 1993; Leggett 1992). The structure of the model, as it pertains to strontium and radium is provided in Figure B.2. Like calcium, the long term repository for both elements is primarily the skeleton, which accounts for 84% and 97% of the effective dose per unit injection of ^{226}Ra and ^{90}Sr , calculated using the ICRP Publication 60 schema (ICRP 1991), respectively.

More recently, alternative models describing the systemic behaviour of strontium and radium have been proposed. Polig et al (2004) developed a model for radium motivated partly by the author's view that there are limitations in the ICRP Publication 67 model as it relates to the deposition and clearance of radium from the skeleton, primarily the absence of a direct link between plasma to bone fluid perfusion of bone volume in the ICRP model; this pathway is included in the model proposed by Polig et al (2004). In the ICRP model this pathway is broadly represented by rapid exchange between bone surfaces and exchangeable bone volume. On the other hand, in the model of Polig et al (2004), bone surfaces are isolated kinetically from the rest of skeleton and represent a site of rapid exchange between bone surfaces and plasma. However, despite these conceptual differences, both models were developed from essentially the same human data and reproduce the observed time dependent retention of radium in human skeleton and soft tissue. Thus any difference in effective dose for ^{226}Ra calculated using the two models will result only from differences in the time dependent deposition of energy in bone surfaces and marrow, which are target tissues in the ICRP Publication 60 schema (ICRP 1991). That this is the case was confirmed by calculations by the present authors, which showed that the effective dose per unit ingestion for adults is around 30% lower when calculated using the model proposed by Polig et al (2004) using the ICRP Publication 60 schema.

Shagina et al (2003) demonstrated significant age and gender specific differences in strontium whole body retention among residents of the Techa River region following exposure, via ingestion, to ^{90}Sr in effluents discharged from the Mayak facility. The data suggest that the ICRP Publication 67 model for strontium generally overestimates skeleton retention, particularly for females, adolescents and older adults (>age 25 years), indicating that the model may give conservative dose estimates for these categories. Better agreement was observed between data and model predictions for adult males, which is consistent with earlier data and a later study by Nair et al (2006), which showed that measurements of strontium in the skeletons of adult male accident victims were consistent with predictions made using the ICRP model. For the purpose of obtaining more accurate estimates of absorbed tissue doses for the epidemiological study of Techa River residents, Shagina et al (2003) proposed modifications to the ICRP Publication 67 model for strontium that allowed for age and gender specific changes in the deposition and turnover of strontium in bone; they termed the revised model the Techa biokinetic model (TBM); note that the ICRP model also allows for age dependent changes in skeleton retention of strontium but only up to age 20 years, and for fewer time steps.

Based on the above, for the present study the age-dependent ICRP Publication 67 models (ICRP 1993) were assumed to provide a reasonable representation of the distribution and retention of strontium and radium in the body for all age groups for uncertainty analysis, noting that age and gender specific skeleton retention may deviate from the central model predictions.

Biokinetic treatment of ²²⁶Ra progeny

²²⁶Ra has a number of short lived radioactive progeny, the most important being the decay series ²²²Rn to ²¹⁴Po, which contributes an additional dose from alpha-decay. Apart from isotopes of radon, the decay products are assumed to follow the behaviour of the radium parent in bone until they are removed from bone volume. Outside the skeleton, they are assumed to follow radionuclide specific biokinetics. The ²²²Rn progeny produced in the lungs, or systemic tissues, from decay of ²²⁶Ra are assumed to be rapidly removed from the body (ICRP 1993). For this reason, it was assumed that uncertainty on doses from the decay of ²²⁶Ra could be adequately represented by considering uncertainty in the structure and parameter values of the ²²⁶Ra parent systemic model alone. Although ICRP is considering a systemic model for radon in its forthcoming publication on occupational intakes of radionuclides, inclusion of this model lay outside the scope of the present analysis.

B2.2 Uranium

The structure of the model used by ICRP (1995b) to describe the biological behaviour of uranium after it has been absorbed to blood from the lungs or the alimentary tract is similar to the generic model developed for the alkali-earth metals (ICRP 1993) and reflects the similar systemic behaviour of the uranyl (UO₂⁺) and Ca²⁺ ions. The model is largely based on three studies of the behaviour of uranium in humans; these are the so-called Boston study (Struxness et al 1956), Bassett study (Bassett et al 1948) and Terepka study (Terepka et al 1964). The studies were all conducted on hospital patients, with the Boston subjects being in the poorest physical condition. This raises the question as to whether the structure of the model is applicable to healthy adults and juveniles. However, the model is supported by additional autopsy data from healthy human subjects at long times after intake (Leggett 1994). In this report, it was assumed that the structure of the model is a realistic representation of the systemic behaviour of uranium in representative members of the public. This assumption has fewer implications for inhalation of moderate to insoluble uranium, where weighted equivalent dose to the lung constitutes around 90% of the effective dose per unit intake.

B2.3 Caesium

Current dose coefficients for members of the public provided in ICRP Publications 71 and 72 (ICRP 1995, 1996) were calculated using a simple two compartment model, which can be represented by the following function:

$$B(t) = A.e^{-\frac{0.693}{T_1}t} + (1 - A)e^{-\frac{0.693}{T_2}t} \quad (B.1)$$

Where $B(t)$ is the whole body retention at time, t , after uptake from blood of inhaled or ingested activity. Values for compartments T_1 and T_2 were derived for adults in ICRP Publication 30 (ICRP 1979), having values of 2 and 110, respectively. It is assumed that 10% of caesium entering the blood from the lungs or alimentary tract is allocated to compartment T_1 , the remainder to T_2 (hence $A=0.1$). The compartments T_1 and T_2 broadly represent the rapid excretion of caesium initially accumulated in the kidneys and longer term excretion of caesium accumulated in muscles and other soft tissues, respectively (ICRP 1989). The model was later extended to children by Leggett (1986), on the basis that a close relationship could be demonstrated between potassium and caesium retention in human subjects of varying age. For 1 year olds it is assumed that all caesium taken up to blood is allocated to compartment T_2 ($A=0$), with a removal half-time from this compartment of 13 days. For 10 year olds, 30% of caesium taken up to blood is allocated to compartment T_1 ($A=0.3$), and the half-times for removal from compartments T_1 and T_2 are 5.8 and 50 days, respectively.

A more realistic physiologically based pharmacokinetic (PBPK) model representing the retention of caesium in systemic tissues has been published by Leggett et al (2003). Here, model parameters and compartments are intended closely to represent actual physiological processes governing uptake and retention in body tissues. PBPK models have the advantage that variability in biological processes governing retention can be represented more accurately for modelling population kinetics (Clewell et al 2008). In the revised model, uptake of caesium from blood to systemic tissues is determined by the fraction of cardiac output perfusing the tissue and the “extraction efficiency” of each tissue in accumulating caesium; this is related to the efficiency of the membrane bound K^+/Na^+ -ATPase in accumulating caesium ions and the number of these transporters in each tissue. The rate of return of caesium from systemic tissues is assumed to be related to cardiac output, the fraction of caesium extracted by individual tissues, and the caesium content of tissues and plasma under equilibrium conditions (Leggett et al 2003). The values of parameters in the proposed model have been calibrated against adult human data. There is currently no similar model for children. An adaptation of the proposed model will be used to calculate dose coefficients for adult workers in the forthcoming publication on occupational intakes of radionuclides to be published by ICRP (ICRP, in preparation). The adapted model is considered in the present analysis. The adaptations simplify the model for use with the source organs considered in ICRP Publication 60 (ICRP 1990). These modifications are as follows:

- The generic soft tissue compartment “Other 1” incorporates the compartments adipose tissue, skin, and brain.
- Compartments spleen, pancreas, and tissues of the heart, lung, and gastrointestinal tract are combined into a generic soft tissue compartment “Other 3”.
- The compartment “Other skeleton” is divided into four groups: “trabecular bone surface”, “trabecular bone volume”, “cortical bone surface”, and “cortical bone volume”. Each group is assumed to receive 25% of the transfer from plasma to “Other skeleton”.

B2.4 Plutonium and americium

The dose coefficients for plutonium and americium bearing compounds for members of the public were calculated using a common model structure derived for bone surface seeking radionuclides in ICRP Publication 67 (ICRP 1993). The model has since been revised for plutonium by Leggett et al (2005) in the light of more recent bioassay data, including autopsy data of former workers of the Mayak Production Association (MPA) published by Suslova et al (2002), and more recent data from a UK volunteer study (Talbot et al 1997). The revised model predicts some differences in the early behaviour of plutonium but gives similar values of effective dose per unit ingestion of plutonium to the ICRP Publication 67 model. Blanchardon et al (2007) proposed a modest revision of the ICRP Publication 67 model for americium based on a re-evaluation of autopsy data for former US plutonium workers. However, Puncher and Harrison (2012b) showed that the proposed revision, which is restricted to changes in the values of some model parameters, rather than a revision of the model structure, has only a small impact on effective dose per unit ingestion of americium. A review of the basis of the ICRP Publication 67 models for plutonium and americium and adaptations of the models for different age groups is provided by ICRP (1993) and Leggett (1985, 1992b).

B2.5 Tritiated water (HTO) and organically bound tritium (OBT)

The dose coefficients for members of the public for intakes of tritiated water (HTO) and organically bound forms of tritium (OBT) were calculated using a simple two compartment model as follows:

$$B(t) = A.e^{-\frac{0.693}{T_1}t} + (1 - A)e^{-\frac{0.693}{T_2}t} \quad (\text{B.2})$$

Where $B(t)$ is the whole body retention at time, t , after uptake from blood of inhaled or ingested activity. The half-life of loss from the T_1 compartment was derived from the daily rate of turnover of water in body tissues; the T_2 compartment is assumed to follow the rate of turnover of carbon in the body. The differences between the biokinetics of HTO and OBT are principally due to differences in the partition of activity between the T_1 and T_2 compartments. For HTO it is assumed that 97% of activity is associated with T_1 (body water) and 3% with organic material; for OBT 50% of activity in blood is partitioned in T_1 , 50% in T_2 . The apportionment is assumed to be age independent (ICRP 1989). The rate of turnover of tritium in the two compartments is assumed to be related to the age dependent rate of turnover of bodily water and carbon. These rates decrease with increasing age (ICRP 1989).

The models were intended as simple compartment models for radiation protection purposes, rather than to provide a realistic PBPK type framework for accurate biokinetic modelling of tritium bearing materials. A recognised weakness of the model is its single compartment representation of the retention of organically bound tritium. Taylor (2003) reviewed data on tritium excretion by individuals exposed to HTO and suggested that the organically bound component can be better represented by a compartment with a 40 day half-time (98%) and a longer term 350 day component (2%). The latter probably represents tritium incorporated into slow turnover structural components. This

compartment was not included in the present analysis due to its small contribution to committed effective dose, per unit intake.

In reality, the retention of OBT is closely dependent on the chemical form of the ingested material. The ICRP model for OBT is intended to represent the turnover of tritium ingested in foodstuffs, rather specific forms of organically bound tritium. Hence, the long term component broadly represents the relative rate of metabolism of the various carbon pools in the body (carbohydrates, lipids and protein). To this end, several authors have proposed alternative models that have a more physiologically based structure and consider the effect of different dietary forms of OBT on retention, excretion and dosimetry of tritium bearing compounds, and the metabolic rate of turnover of the different carbon pools (Whillans 2003; Richardson and Dunford 2003a, 2003b; Galeriu and Melintescu 2010).

B2.6 Iodine

The dose coefficients for inhalation and ingestion of iodine were calculated using a simple three compartment model, consisting of blood, soft tissues (excluding thyroid) and thyroid gland; with recycling of material occurring between thyroid and blood (ICRP 1989). This model is based on an earlier one derived by Riggs (1952), and was extended by ICRP (1989) to incorporate age dependent differences in uptake and retention of iodine by the thyroid. A simple model structure is justified for radiation protection purposes on the basis that doses to the thyroid account for nearly 100% of the effective dose resulting from ingestion or inhalation of soluble iodine bearing materials. However, the model has recently been revised for adults by Leggett (2010), to include additional soft tissue compartments and a more detailed representation of iodine uptake and retention by the thyroid gland. The revised model predicts almost identical doses to the thyroid and effective dose per unit intake for longer lived isotopes of iodine, including ^{131}I , ^{129}I and ^{125}I ; but greater discrepancies are observed for short lived isotopes and radioiodine labelled thyroid hormones (Leggett 2010).

The fraction of iodine taken up by the thyroid from blood in 24 hours (the U_{24} value), and hence dose to the thyroid and effective dose, from inhaled or ingested iodine, is inversely correlated with background dietary intakes of iodine; this varies significantly between populations (WHO 2007). Empirical relationships between dietary intake and thyroid uptake of radioiodine have been derived (Zvonova 1989; Leggett 2010). This study considered the effect of dietary intakes of iodine on thyroid doses for a UK population.

APPENDIX C

Derivation of uncertainties

C1 SENSITIVITY ANALYSES

A sensitivity analysis was conducted to guide the derivation of uncertainties in the biokinetic models describing the uptake and retention of each radionuclide. A method described by Kursheed and Fell (1997) was adapted to identify biokinetic parameters in the HRTM, alimentary tract model and appropriate systemic models, having the greatest impact on committed effective dose, E(50) (to 50y) following inhalation or ingestion of 1 Bq. The numerical algorithm calculates the sensitivity coefficient for each rate using the following equation:

$$S_{i,j} = \frac{\Delta D}{\Delta \lambda_{i,j}} \cdot \frac{\lambda_{i,j}}{D} \quad (\text{C.1})$$

Where

- $S_{i,j}$ The sensitivity coefficient for the rate from compartment i to j , with respect to the dose of interest, D .
- D The dose predicted by the model, with all rates set at the values defined in the model.
- $\lambda_{i,j}$ The rate specified in the model from compartment i to compartment j .
- ΔD The observed change in the dose of interest due to the observed change in rate $\lambda_{i,j}$, $\Delta \lambda_{i,j}$.
- $\Delta \lambda_{i,j}$ The change in rate from compartment i to compartment j . For the present analysis, the value of $\Delta \lambda_{i,j}$ is chosen so that $\Delta \lambda_{i,j} / \lambda_{i,j}$ is 0.01.

The sensitivity coefficient is the ratio of the fractional change in dose: fractional increase in the parameter of interest; thus a value of $S=1$ means that a 1% change in the parameter of interest produces a 1% change in the quantity of interest. The coefficients resulting from a typical analysis are provided in Table C.1 for ingestion and inhalation of radium. For intakes of all radionuclides by ingestion, the coefficient for the fraction of activity absorbed from the alimentary tract, the f_1 value, is always close to unity, reflecting the fact that effective dose from ingestion is almost linearly related to this parameter. For intakes by inhalation, the coefficients were greatest for the parameters affecting the amount of activity deposited in the lung, and the overall rate of clearance to blood (dissolution and absorption to blood) and to the alimentary tract (particle transport clearance).

Sensitivity coefficients offer a simple means to identify important parameters in the model that affect the output of interest (in this case, doses). However, such an analysis is local and so does not consider any synergism between model parameters; nor does it

consider the degree of uncertainty (variance) on values of the parameters. The sensitivity analyses were therefore used to assist in the model parameters that were chosen to vary.

C2 UNCERTAINTIES IN THE ALIMENTARY TRACT MODEL

C2.1 Uncertainty in the fraction of material absorbed to blood from the alimentary tract: the f_1 value

With the exception of tritiated water, whose rate of absorption to blood was assumed to be instantaneous, distributions were derived for the f_1 parameter for each radionuclide. The derived distributions were assumed to represent uncertainty in the mean population value for the stated age group: 1 year olds, 10 year olds and adults. The derived distributions are given in Table C.2.

Uranium

The amount of uranium absorbed from the alimentary tract depends on individual dietary habits, nutritional status and age. Data relating to the absorption of uranium from the alimentary tract have been reviewed by ICRP (1995b), Leggett and Harrison (1995), and later by Harrison et al (2001). For ingestion of uranium in drinking water, which probably represents the most likely route of environmental exposure to uranium, the f_1 values ranged from 0.0025 to 0.06, with central values of 0.01-0.02. Harrison et al (2001) suggested a 90% range of f_1 values, for ingestion of uranium by members of the public, of between 0.006 and 0.03 for adults, and 0.008 to 0.05 for 10 year old children. The ranges were derived using formal expert elicitation techniques, and the uncertainty on f_1 was expressed as a subjective probability statement regarding the 90% probability interval of the true *mean* value of f_1 for ingested uranium compounds.

Alexandrou (2010) fitted a lognormal distribution to the f_1 values obtained from a later study by Zamora et al (2002). This study measured the absorption to blood of uranium ingested in food and water by Canadian members of the public who varied in age from 13 to 87 years. The fitted distribution had a median value of 0.01 and GSD of 2.4; with a range of 0.002-0.04, which is consistent with the results of earlier studies.

In this report, the distribution derived for the f_1 parameter is assumed to represent uncertainty on the *population mean* values. For adults the uncertainty was assumed to be represented by a lognormal distribution with median equal to the ICRP value (0.02) and GSD of 1.6, which gives a 90% range of values of 0.009 to 0.04. For 1 and 10 year old children a lognormal distribution with median value of 0.02 and GSD of 1.8 was assumed, which gives a 90% range of 0.008 to 0.05.

Strontium

The data relating to the absorption of strontium from the alimentary tract has been reviewed by Harrison et al (2001), who suggested that uncertainty on the fraction of strontium absorbed to blood from the alimentary tract, the f_1 value, could be represented by a range of 0.1-0.4 for adults, and 0.1-0.5 for children. These ranges are consistent with a later review by Apostoaei (2002) and a study by Li et al (2006), which suggest f_1 values for adults of around 0.2 and 0.5, respectively. For this analysis, uncertainty in the

f_1 value was modelled using a triangular distribution with minimum value of 0.1, maximum value of 0.3 and mode (vertex of the triangle) of 0.3. For 1 and 10 year old children a triangular distribution was assumed with minimum value of 0.1, maximum value of 0.5 and mode of 0.4 (the ICRP value for children age 1 year or older).

Radium

The data relating to the absorption of radium from the alimentary tract has been reviewed by Harrison et al (2001), who suggested ranges of f_1 values of 0.05 to 0.3 for adults and 0.05 to 0.4 for 10 year old children; the review suggests that the ICRP values of 0.2 for adults and 0.3 for children are likely close to the population mean values for these age groups. In this study, uncertainty in the f_1 value was modelled using a triangular distribution with minimum value of 0.05, maximum value of 0.3 and mode (vertex of the triangle) of 0.2. For 1 and 10 year old children a triangular distribution was assumed with minimum value of 0.05, maximum value of 0.4 and mode of 0.3 (the ICRP value for children age 1 year or older).

Caesium

The data relating to the absorption of caesium from the alimentary tract after ingestion of caesium in food has been reviewed by Harrison et al (2001), who suggested a 90% coverage probability of 0.8 to 1.0. Based on this, uncertainty on the f_1 value was assumed to be adequately represented by a uniform distribution with lower bound of 0.8 and upper bound of unity.

Plutonium

The data relating to the absorption of plutonium from the alimentary tract has been reviewed by Harrison et al (2001), who suggested a range of f_1 values, for non-specific plutonium bearing compounds ingested by members of the public (all age groups), of between 1×10^{-4} and 1×10^{-3} , and median value of 5×10^{-4} . This range was derived using formal expert elicitation techniques, and the uncertainty on f_1 was expressed as a subjective probability statement regarding the 90% confidence interval of the true mean value of f_1 for non-specified plutonium compounds. Based on the review and distribution derived by Harrison et al (2001), uncertainty on the mean value of f_1 , for plutonium was expressed as a log-normal distribution with median of 5×10^{-4} and GSD of 1.73; this gives a 90% probability interval of 2×10^{-4} - 1.2×10^{-3} .

Americium

Absorption of americium from the alimentary tract has been less extensively studied than absorption of plutonium. ICRP assigns an f_1 value of 5×10^{-4} for members of the public (ICRP 1996), the same as plutonium; furthermore, where estimates of f_1 values have been made for both radionuclides in shellfish, those for americium appear consistent with the ranges observed for plutonium (Hunt et al 1986, 1990; Hunt 1998). For this analysis, uncertainty on the f_1 value for americium was assumed to be the same as that for plutonium.

Iodine

The data relating to the absorption of iodine from the alimentary tract has been reviewed by Harrison et al (2001), who suggested a 90% confidence interval of 0.9-1.0 for all members of the public. Because existing data suggest that the f_1 value for iodine in food is likely to be closer to unity, uncertainty on the population mean value was assumed to be represented by a triangular distribution with minimum of 0.9 and maximum value and vertex of unity.

Organically Bound Tritium (OBT)

The distribution of the f_1 value for OBT derived by Harrison et al (2002) was assumed. This is a uniform distribution with lower bound of 0.9 and upper bound of unity.

C3 UNCERTAINTIES IN THE HUMAN RESPIRATORY TRACT MODEL

With the exception of exposure to iodine and tritiated water, which were assumed to be inhaled in vapour form, exposures via inhalation were assumed to occur via inhalation of poly-disperse aerosols.

Sensitivity analysis suggested that only a few HRTM parameters are significant, including those governing long term clearance from the lungs. This is consistent with previous studies (Puncher et al 2008; 2011) showing that the lung clearance parameters, in particular the slow dissolution rate s_s significantly affect the uncertainty on dose determined from urine bioassay. However, the study by Puncher et al (2011) showed that collectively, the other HRTM parameters, in particular those that determine regional deposition in the lungs, were also important. In the present study therefore, it was considered appropriate to derive distributions for the principal lung deposition and clearance parameters as considered by Puncher et al (2011). The distributions are summarised in Table C.3.

C3.1 Deposition of iodine gas

It was considered that exposures via inhalation to iodine in the UK occur primarily via inhalation of iodine vapour, most likely in the form of methyl iodide gas (CH_3I) (Collins et al 2004) released as a result of nuclear power generation (^{131}I) and fuel reprocessing (^{129}I). In ICRP Publication 71 (ICRP 1995a) it is assumed that methyl iodide behaves as a class V-1 vapour: 70% of the inhaled vapour is assumed to be deposited in the thoracic and extra-thoracic airways (ET_1 0%; ET_2 21%; BB 7%; bb 14%; Al 28%), and absorbed instantaneously to blood. Thus there is a 100% correlation between the fraction deposited in the airways and effective dose per Bq of inhaled radioiodine. Morgan et al (1967) performed studies on human volunteers exposed to methyl iodide gas which suggested that the amount deposited varied between 50-90%, with a mean value of 70%. For the present study, uncertainty on the mean value was assumed to follow a normal distribution with mean of 70% and standard deviation of 15% (which gives a 95% range of values of 40-100%); this is likely to overestimate uncertainty on the population mean value but accounts for the possibility that the chemical forms may include other iodine species in addition to methyl iodide. Uncertainty in the fraction

deposited was implemented by scaling the regional deposition fraction of 70% by a common factor, V_{DP} , sampled from a normal distribution with mean of unity and standard deviation of 0.21.

C3.2 Aerosol activity median aerodynamic diameter (AMAD) and aerosol geometric standard deviation (GSD)

Environmental poly-disperse aerosols can be conveniently described by a lognormal distribution defined by an activity median aerodynamic diameter (AMAD): 50% of the radioactivity is associated with particles having an aerodynamic equivalent diameter greater than the AMAD; and geometric standard deviation (GSD).

Measurements of environmental aerosols have been reviewed by Dorrian (1997), who compiled nearly 200 measurements of AMAD values from aerosols resulting from the Chernobyl incident, fallout from nuclear weapons tests, and aerosols present around nuclear facilities. The aerosols covered a range of materials and radionuclides. The range of AMAD values could be described by a lognormal distribution with median value of 1.5 and GSD of 2.9; the GSD values of the aerosol distributions followed a lognormal distribution with median of 2.5 and GSD of 1.3. The distributions derived for aerosol AMAD and GSD by Dorrian (1997) were assumed for each analysis where exposure was assumed to occur via inhalation of polydisperse aerosols (all exposures except inhalation of iodine). It was assumed that the AMAD and GSD values were uncorrelated.

C3.3 Breathing parameters

The primary breathing parameters are the ventilation rate, B ($\text{m}^3 \text{h}^{-1}$), breathing frequency, f_R (breaths per minute) and tidal volume, V_T (Litres). Bailey et al (1997) estimated that the mean ventilation rate among adult males varied by a factor of within 2 for different levels of exercise (sleep, sitting, light exercise, heavy exercise). This can be represented by a lognormal distribution with median value equal to the ICRP Publication 66 value (ICRP 199ab), and GSD of 1.3. In this analysis the ventilation rate, B , of each reference level of exercise for a member of the public (ICRP 1995a) was varied using a common factor with median of unity and GSD of 1.3; the rates were therefore assumed to be 100% correlated. The respiratory frequency for each level of exercise, f_R , was assumed to be 100% correlated with the ventilation rate and also followed a lognormal distribution with median equal to the ICRP reference value and GSD of 1.3.

C3.4 Fraction breathed through the nose, F_n

The distribution for the fraction breathed through the nose was based on the findings of Niinimaa et al (1980, 1981) who monitored the switch from nose to mouth breathing among 30 volunteers during the transition from light to heavy exercise. Based on the frequency of occurrence of volunteers classed as nose breathers, mouth breathers or normal augmenters, in the Niinimaa studies, and assuming the average percentage time spent in the activities sleeping, sitting and light and heavy exercise suggested by ICRP for a reference member of the public (ICRP 1995a), a right angled triangular

distribution PDF with minimum value of 0.4, and vertex and maximum value of 1 was assumed.

C3.5 Particle transport clearance

The reference particle transport clearance rates were derived by ICRP from a number of sources including animal studies and involved an element of judgment. The values were considered to represent central or more precisely, median values in the human population (M. R. Bailey, personal communication). ICRP assumes that inter-subject variation or variability in particle transport clearance varies by a factor of three around the reference value, and that this can be represented by a lognormal distribution with median equal to the ICRP reference value and GSD of 1.73 (ICRP 1994b). In an uncertainty analysis of lung doses from inhaled Depleted Uranium (DU), Puncher et al (2008) varied all particle transport rates in the HRTM by the same factor, K_{PT} , where K_{PT} was randomly generated from this distribution. Thus it was assumed that all rates were 100% correlated, because lung doses from inhaled DU appear to be sensitive to only a few of the particle transport clearance rates (Bailey and Puncher 2007). However, the movement of material from the alveolar-interstitial (AI) region to the bronchiolar region and thoracic lymph nodes occur by very different processes from those governing transport from the bronchiolar and bronchial airways. The former are facilitated by macrophages in the alveolar-interstitial (AI) region, the latter by mucociliary transport along airways walls. For the revised model of clearance from the AI region given in Figure B.1, Gregoratto et al (2010) suggest that variability in the rate of clearance from the alveolar region to the bronchiolar region (the pathway ALV to bb1 in Figure B.1) is more accurately described by a lognormal distribution with median of 0.001 and GSD of 4.5, and the pathway of clearance from the alveolar region to the interstitium (INT) by a lognormal distribution with median of 0.0013 and GSD of 3.2. Being so low, the rate from INT to the thoracic lymph nodes, LN_{TH} , is also very uncertain. In a Bayesian analysis of plutonium workers, Puncher et al (2011) assumed that variability in this rate could be represented by a lognormal distribution with median of 3×10^{-5} and GSD of 3. The same distribution was assumed in this analysis. The remaining rates were varied by a common factor K_{PT} , where K_{PT} was randomly sampled from a lognormal distribution with median of unity and GSD of 1.73.

C3.6 Absorption parameters

In the HRTM (ICRP 1994b), the absorption to blood of material deposited in the extra thoracic region (ET_2) and the thoracic airways is assumed to occur by a two-stage process:

Dissolution of the inhaled material. This is described by three parameters: the fraction of material that dissolves rapidly, f_r , the dissolution rate of the rapid fraction, s_r , and the dissolution of the remaining fraction ($1 - f_r$), at a rate s_s .

Uptake of dissolved material to blood. This is assumed to be instantaneous unless the dissolved ions of the radionuclide become bound to the airway walls. The proportion of material that becomes bound after dissolution is represented by the bound fraction, f_b . Since the ionic form of the radionuclide determines the extent of any bound state, it is

assumed to be independent of the chemical form of the inhaled material. Uptake of material to blood from the bound state is assumed to occur at a rate, s_b . For the radionuclides in this report, with the exception of plutonium (discussed below), existing experimental data reviewed by ICRP suggests little or no binding in the respiratory tract (ICRP 2002). In this analysis therefore, it was also assumed that $f_b = 0$.

The absorption parameter values are assumed to be material specific and, with the exception of uranium, are assumed to represent the lung solubility of a particular material, and so represent a source of uncertainty.

Uranium

The values of the dissolution parameters are dependent on the chemical form of the inhaled material. ICRP reviewed solubility parameters for a range of uranium bearing materials in Publication 71; studies of common chemical forms suggest that uranium compounds fall across the spectrum of Type F (fast) to Type S (slow) absorption types. ICRP suggests assuming the default Type M (medium) absorption type when the chemical form is uncertain. A review of environmental forms of uranium (Newsome, personal communication), suggests that environmental uranium aerosols are more likely to be insoluble oxide forms. In this analysis it was assumed that uncertainty on the chemical form was likely to be the principal source of uncertainty. Puncher et al (2011) reviewed available *in vivo* data describing the lung solubility of various chemical forms of uranium and derived distributions that are inclined towards more insoluble forms. The distributions for the dissolution parameters (f_r , s_r and s_s) derived by Puncher et al (2011) were used in the present analysis since they were deemed reasonable to represent uncertainty about the chemical form of uranium compounds inhaled in the environment.

Caesium and strontium

Exposures by inhalation to environmental forms of caesium and strontium are attributable to inhalation of residual aerosols produced from reactor accidents, principally the Chernobyl incident in 1986, and also to fallout aerosols produced by atmospheric nuclear tests conducted in the 1940s to early 1960s. Studies of the solubility characteristics of these aerosols suggest mainly soluble forms. Studies on Chernobyl aerosols suggest values of the rapid fraction, f_r , of around 0.04 to 0.4, with a mean value of between 0.1-0.3 (Kutkov 1998, 2000; Cuddihy 1989; Kutkov and Komiritskaya 1986). Particulates of caesium bearing aerosols resulting from nuclear power generation support values of between 0.4-0.8 (Stradling et al 1996, 1997; Dua et al 1987). Collectively, these studies suggest values of the slow dissolution rate of between ~ 0.001 - 0.005 d^{-1} . For this study, uncertainty on the value of the rapid fraction was assumed to be represented by a triangular distribution with minimum value of zero, maximum value of unity and vertex of 0.21. The rapid rate was fixed at 3 d^{-1} , the default value for strontium compounds being considered by ICRP, based on a review of strontium bearing materials (ICRP, in preparation). Uncertainty on the slow dissolution rate was assumed to be represented by a uniform distribution with lower bound of 0.001 d^{-1} and upper bound of 0.005 d^{-1} .

Radium aerosols

Very few studies have attempted to derive lung absorption parameter values for radionuclides embedded in fly ash. However, it seems reasonable to assume that these will be relatively insoluble given that fly ash consists mostly of silicon dioxide in the form of quartz or amorphous silica. Kalkwarf et al (1984) performed an *in vitro* study to determine dissolution rates for uranium, radium and other radionuclides from fly ash samples using simulated lung fluid. The solubility of the uranium component was variable, with a rapid dissolution fraction of between 2 and 20% and slow dissolution rate of between 1×10^{-3} and $6 \times 10^{-5} \text{ d}^{-1}$. For other radionuclides, which included ^{226}Ra , less than 0.2% had dissolved during the course of the trial. This could be interpreted as meaning either 0.2% of the material was soluble ($f_r = 0.002$; $s_r > 0.2$; $s_s = 0 \text{ d}^{-1}$), or that 100% of the material dissolves at a rate of around $3 \times 10^{-4} \text{ d}^{-1}$ ($f_r = 1$; $s_s = 3 \times 10^{-4} \text{ d}^{-1}$), or, more than likely, some combination of these extremes. However, the short duration of the study means that the accurate identification of parameter values, particularly the slow dissolution rate, is subject to a large degree of uncertainty. Furthermore, there is also considerable uncertainty in extrapolating the *in vitro* behaviour of the material to the lungs. Nevertheless, assuming either set of parameters inferred from the study of Kalkwarf et al (1984) assigns fly ash Type S (slow solubility) behaviour according to ICRP criteria (ICRP 1995a).

Griffis and Snipes (1981) performed an *in vivo* study to determine the overall clearance of radionuclides present in neutron activated fly ash from rat lungs at times up to 120 days post inhalation. The rate of clearance varied depending on the radionuclide: the clearance of ^{134}Cs was around five times faster than ^{59}Fe , the latter being highly insoluble. However, inferring values of the lung dissolution parameters is complicated by uncertainties in the rate of particle transport clearance from the rat lung and the method the authors used to determine the thoracic content of radionuclides *in vivo*.

The choice of appropriate distributions to represent uncertainty on the lung dissolution parameters of fly ash is difficult given the paucity of data. Nevertheless, these suggest that radium in fly ash is perhaps more likely to exhibit Type S (slow solubility) than Type M (medium solubility) behaviour.

Given the above, it was assumed that uncertainty on the rapid fraction, f_r , could be represented by a log-uniform distribution with lower bound value of 0.001 and upper bound value of 0.1; the slow dissolution rate, s_s , by a log-uniform distribution with lower bound of 1×10^{-5} and upper bound of $5 \times 10^{-3} \text{ d}^{-1}$. The bounds of these distributions are consistent with the study of Kalkwarf et al (1984), but also broadly cover the range of parameter values that have been collated for a variety of radionuclide bearing materials that can be classed as Type M or S (ICRP, in preparation). The choice of a log-uniform distribution is consistent with the belief that the material is "as likely" to be insoluble as moderately soluble. The use of a log-uniform distribution also avoids the need to specify a modal value. It was assumed that the rate of the rapid fraction, s_r , was 3 d^{-1} , which is the default value being considered for Type M and S materials in the forthcoming ICRP publication on occupational intakes of radionuclides (OIR) (ICRP, in preparation).

Plutonium aerosols

It was assumed that environmental plutonium bearing aerosols were likely to be in the form of insoluble oxides. Values of absorption parameters for plutonium oxides determined from *in vivo* studies of animals and humans have been collated by Puncher et al (2011), who fitted lognormal distributions to the rapid fraction, f_r , rapid dissolution rate, s_r , and slow dissolution rate s_s . These distributions were used to estimate uncertainties on lung doses for plutonium workers exposed to plutonium bearing aerosols. The same distributions were assumed adequately to represent the dissolution characteristics of environmental plutonium bearing aerosols in this study. The analysis of Puncher et al (2011) also included distributions for the parameters that determine long term binding in the lungs. In the present analysis a bound state was not included because preliminary analyses indicated that the values of the bound state parameters considered by Puncher et al (2011) have an almost negligible effect on the distribution of effective dose per unit inhalation of plutonium oxide.

Americium aerosols

A review by ICRP suggests that inhalation exposures to environmental americium will most likely occur via association of americium with plutonium bearing aerosols, and therefore share the dissolution characteristics of the plutonium matrix with which it is associated. This assumption appears to be supported by *in vivo* studies of rats exposed to plutonium oxide bearing particulates (ICRP 1995a). For this study, the distributions for the dissolution parameters assumed for plutonium bearing aerosols were assumed for americium. In the absence of evidence to the contrary, it was also assumed that no long term binding of americium occurs in the airways.

C4 UNCERTAINTIES IN SYSTEMIC MODELS

C4.1 Uranium

The decision on which rate constants to vary was guided by a local sensitivity analysis of effective dose per unit ingestion of uranium; and consideration of the weighted equivalent organ doses that make the greatest contribution to effective dose among systemic tissues. These were the rates governing uptake and retention by the skeleton and liver, the rate of uptake to the compartment representing long term soft tissue retention (compartment "ST2" in the model described by Leggett 1994), and the rate of loss from blood to urine bladder. The derived distributions are summarised in Table C.4.

Data describing the biological behaviour of uranium in humans and animals has been reviewed by Leggett (1994) and later by Harrison et al (2007). The data are relatively sparse, making it difficult to distinguish uncertainty on the central or reference values in the model from inter-subject variation (variability) in the systemic behaviour of uranium. Where possible, the chosen approach was to select distributions for rate constants that reproduce the range of organ content observed in the available human dataset. The selection of distributions was also constrained to ensure that the median values of the distributions of simulated bioassay predictions were close to those calculated using the unperturbed model at different times post intake; the chosen distributions gave

agreement between median values of simulated bioassay quantities and unperturbed model values of typically within 5%.

Urinary excretion

The inter-subject range of urinary excretion of uranium can be achieved by varying the rate from blood to bladder by a factor of three above or below the reference model value (Harrison et al 2007). This was achieved by multiplying the rate by a random variable generated from a lognormal distribution with median value equal to unity and geometric standard deviation (GSD) of 1.73. This sampling regime reproduces the inter-subject variation in cumulative urinary excretion observed in the human dataset: ~53-89% of injected activity (95% range), one week post injection.

Uptake to the skeleton and liver from blood

All rates to bone surfaces from blood were varied together (assumed to be 100% correlated) using a common scaling factor that was randomly generated from a lognormal distribution with a median of unity and GSD of 1.3; the rate from blood to liver was varied using a lognormal distribution with median equal to the model value and GSD of 1.4. This sampling regime reproduced the following:

Inter-subject variation (the inter-quantile $Q_{0.025}$ - $Q_{0.975}$ range) in simulated liver retention at times up to 1000 days post-injection was around a factor of 10, consistent with the ranges observed in human subjects collated by Leggett (1994).

The ratio of simulated skeleton:liver content at 10,000 days post injection was consistent with the range of post-mortem measurements of skeleton:liver content determined from healthy occupationally and non-occupationally exposed individuals (Leggett 1994). The observed ratios are well represented by a lognormal distribution with median of 0.34 and GSD of 1.9 ($P < 0.24$); the difference between this distribution and the simulated distribution is not statistically significant ($P < 0.7$).

The simulated skeleton retention at one year was 1-10% of the injected activity, in accordance with a previous study by Harrison et al (2007).

Rates within skeleton

Rates within skeleton were varied in accordance with a previous study by Harrison et al (2007). All rates from bone surfaces were varied together (assumed 100% correlated) by a factor of 9 above and below the central value; this regime was implemented by varying rates from bone surfaces together using a common factor sampled from a lognormal distribution with a median of unity and GSD of 3.

All rates from bone volume were varied by a factor of 2 above and below the model values. Because the mechanism of turnover from exchangeable bone volume is different from non-exchangeable bone volume, variation in the rates from exchangeable bone volume and non-exchangeable volume were assumed to be uncorrelated. This regime was implemented by varying rates from exchangeable volume together using a common factor sampled from a lognormal distribution with a median of unity and GSD of 1.4; rates from non-exchangeable bone volume were varied using a common factor sampled from a lognormal distribution with median value of unity and GSD of 1.4. This

range is consistent with observed variation in plasma concentration of biochemical markers for bone resorption in adults (Fatayerji and Eastell 1999, Nguyen et al 2007, Eastell et al 1988).

Long-term retention in massive soft tissues

The rate of uptake of uranium from blood to massive soft tissue (compartment "ST2" in the model described by Leggett 1994) was varied using a lognormal distribution with median equal to the model value and a GSD of 1.73. This produced a geometric inter-quantile $Q_{0.025}$ - $Q_{0.975}$ range in soft tissue content of around a factor of 15, from one year to 50 years post-injection, which is consistent with the ranges observed in the human data up to 600 days post-injection (Leggett 1994).

C4.2 Strontium and radium

The decision on which rate constants to vary was guided by a local sensitivity analysis, conducted using a numerical method described elsewhere (Puncher and Harrison 2012b), to determine which systemic transfer rates have the greatest impact on integrated effective dose per unit ingestion of ^{90}Sr and ^{226}Ra . In addition to the fraction of activity absorbed to blood from the alimentary tract, (the f_1 value), the results indicated that the dose for both radionuclides is most sensitive to variation in the rates governing deposition and removal of activity in skeleton, consistent with the fact that doses to skeleton constitute a large fraction of the effective dose per unit ingestion. A key difference was that, for radium, dose was sensitive to the rate of transfer from blood to colon, but insensitive to the rate of transfer from blood to bladder; with the opposite being true for strontium. These results are consistent with the fact that transfer from blood to colon is the main excretion pathway for radium, but excretion of activity in urine is the main excretion pathway for strontium. The derived distributions are summarised in Table C.5.

Data describing the biological behaviour of radium and strontium in humans and animals have been reviewed by Leggett (1992a) and ICRP (1993), and later by Apostoaei and Miller (2004) for strontium. The data are relatively sparse, making it difficult to distinguish uncertainty on the central or reference values in the models from inter-subject variation (variability) in systemic behaviour. Where possible, the chosen approach was to select distributions for rate constants that reproduce the variation in organ content observed in the available human dataset. In support of this approach, it was found that applying virtually the same sampling regime gave good agreement between the calculated variation in model predictions and data, for strontium and radium. The selection of distributions was also constrained to ensure that the median values of the distributions of simulated bioassay predictions were consistent with those predicted by the model (with reference values for transfer rates) at different times post intake.

Excretion

Variation in the rate of excretion from blood to colon (radium and strontium) and from blood to urine bladder (strontium only) was modelled by multiplying the rate(s) by a random variable generated from a lognormal distribution with median equal to unity and

geometric standard deviation (GSD) of 1.6; thus variation in the excretion pathways for strontium was assumed to be correlated. This regime reproduces the inter-subject variation in whole body retention of radium and strontium inferred from data reviewed in the literature (Leggett 1992a; ICRP 1993; Apostoaei and Miller 2004).

Retention by the skeleton

All rates to bone surfaces from blood were varied together (assumed to be 100% correlated) using a common scaling factor that was randomly generated from a lognormal distribution with a median of unity and GSD of 1.3; this sampling regime reproduced inter-subject variation in the skeletal retention of radium inferred from the limited studies of radium retention in human subjects, principally the study of Schlenker et al (1982), and variation in whole body retention of strontium in the values collated by Apostoaei and Miller (2004).

Rates within skeleton

Rates within skeleton were varied using a regime derived for uranium (a physiological homologue of radium and strontium) in a previous uncertainty analysis of the ICRP systemic model undertaken by Harrison et al (2007). All rates from bone surfaces were varied together (assumed 100% correlated) by a factor of 9 above and below the central value; this regime was implemented by varying rates from bone surfaces together using a common factor sampled from a lognormal distribution with a median of unity and GSD of 3.

All rates from bone volume were varied by a factor of 2 above and below the model values. Because the mechanism of turnover from exchangeable bone volume is different from non-exchangeable bone volume, variation in the rates from exchangeable bone volume and non-exchangeable volume were assumed to be uncorrelated. This regime was implemented by varying rates from exchangeable volume together using a common factor sampled from a lognormal distribution with a median of unity and GSD of 1.4; rates from non-exchangeable bone volume were varied using a common factor sampled from a lognormal distribution with median value of unity and GSD of 1.4. This range is consistent with observed variation in plasma concentration of biochemical markers for bone resorption in adults (Fatayerji and Eastell 1999, Nguyen et al 2007, Eastell et al 1988).

Long-term retention in massive soft tissues

The rate of uptake of radium and strontium from blood to massive soft tissue compartment ST2 was varied using a lognormal distribution with median equal to the model value and a GSD of 1.6. This rate, in conjunction with the regime selected to model variation in excretion, produced a geometric 95% range in soft tissue content of around a factor of 10, up to 30 years post injection, which is consistent with the ranges reported in human subjects.

C4.3 Caesium

A parameter uncertainty analysis of the ICRP Publication 56 model for caesium has been undertaken by Apostoaei and Miller (2004). In their approach, the authors used

the relationship between total potassium in the body, and the model parameters A , T_1 and T_2 given in equation (B.1), derived by Leggett (1986), to propagate variability in potassium content to the calculation of dose per unit ingestion of ^{137}Cs by adults. In the present analysis, the distributions of values for A , T_1 and T_2 , obtained by Apostoaei and Miller (2004) were approximated by lognormal distributions (Table C.6), and the parameters were assumed to be uncorrelated. This sampling regime was justified on the basis that:

1. A local sensitivity analysis of effective dose per unit injection, conducted using the method described by Puncher and Harrison (2012a), indicated that effective dose per unit injection was only significantly sensitive to the parameter T_2 . This was confirmed in a simulation of effective dose per unit ingestion, where only the parameter T_2 was varied using the regime given in Table C.6 and all other parameters were fixed at their median values. In this simulation, the location and variance of the distribution of dose was very similar to the analysis where all parameters were varied.
2. The sampling regime reproduced the observed variability in whole body retention of caesium reported by Leggett et al (2003), and the median value of the distributions of whole body retention at different times were near (typically within 5% of) the values predicted by the model.

Given the paucity of available data describing the retention of caesium in children, it was assumed that the geometric range of the variability in the model parameters derived for adults could be applied to 1 and 10 year old children. Therefore, to calculate uncertainties on doses for these age groups, the model parameters, A (10 year olds only), the rates from compartments T_1 (10 year olds only) and T_2 were multiplied by lognormally distributed random variables with a median of unity and GSDs given in Table C.6.

The effect of uncertainty on model structure was also investigated by performing a parameter uncertainty analysis on the revised systemic model for caesium described by Leggett et al (2003) for adults, or rather the slightly modified version of the model that is compatible with the ICRP Publication 60 and Publication 103 dosimetry framework (ICRP 1991, 2007). A local sensitivity analysis indicated that the effective dose per unit injection was most sensitive to the rates to and from muscle and kidneys to blood, and from kidney to bladder. However, the rates to and from blood to all soft tissues are positively correlated through their relationship between cardiac output and the efficiency of tissues in extracting caesium from plasma via the membrane bound K^+/Na^+ -ATPase enzyme complex. Thus the rates to and from all soft tissues were multiplied by a common factor, B , sampled from a lognormal distribution with median of unity and GSD of 1.4. This regime was chosen because it reproduced the variability in whole body retention of caesium reported by Leggett et al (2003). The choice of sampling regime was further supported by a Bayesian analysis of whole body retention of ^{137}Cs in an adult worker following inhalation exposure to a caesium bearing aerosol, using the revised model and the distribution derived for factor B , as a prior distribution; this gave a posterior distribution of factor B that was well within the 95% range of the prior (Puncher, unpublished).

C4.4 Plutonium

An uncertainty analysis of doses to adult members of the public, resulting from ingestion of plutonium, has been described by Puncher and Harrison (2012b). That study used the revised systemic model for plutonium derived by Leggett et al (2005) for adult workers. To estimate the uncertainties on doses for all age groups in this study, the sampling regime derived by Puncher and Harrison (2012b) was applied to the ICRP Publication 67 model for plutonium (Table C.7). However, unlike the study of Puncher and Harrison (2012b) the rate from blood to liver was assumed not to be negatively correlated with the rate from blood to skeleton. This assumption was made because the rate of plutonium uptake to bone surfaces is assumed to be much higher in 1 and 10 year olds, compared with adults, and the relationship between skeleton and liver uptake derived by Puncher and Harrison (2012b) predicts rates to liver that are less than zero for these age groups. It was found that this modification:

- Did not significantly affect the time dependent variability in skeleton and liver content modelled by Puncher and Harrison (2012b) using the revised model (weighted equivalent doses to these tissues make the greatest contribution to effective dose from the doses to systemic tissues).
- Gave similar distributions of effective dose per unit ingestion for adults calculated using either the ICRP Publication 67 model or revised model for plutonium; the latter were indistinguishable from the distributions calculated by Puncher and Harrison (2012b) using the revised model where the rates were assumed to be negatively correlated.
- Gave better agreement between the simulated liver retention using the ICRP Publication 67 model at late times and the available autopsy data collated by Puncher and Harrison (2012b). This is because the ICRP Publication 67 model predicts significantly higher transfer of activity from blood to skeleton than the revised model: 62% of the total activity transferred from blood to the skeleton and liver is transferred to the skeleton in the ICRP Publication 67 model, compared with 33% for the revised model. The sampling regimes assumed for the ICRP Publication 67 model and the revised model are given in Tables C.7 and C.8 respectively.

C4.5 Americium

An uncertainty analysis of doses from ingestion of americium by members of the public, using the ICRP Publication 67 model for americium (ICRP 1993), has been performed by Puncher and Harrison (2012b). The same sampling regime was applied here to calculate uncertainties on doses from ingestion and inhalation for all age groups. However, in this study it was assumed that the rate from blood to liver was not negatively correlated with the rate from blood to skeleton, as was assumed in the analysis of Puncher and Harrison (2012b), and for the same reasons given for plutonium (above). It was found that assuming that the rates were uncorrelated had a very small effect on the simulated variability in skeleton and liver retention, and the distribution of effective dose per unit ingestion, compared with the distributions calculated in the analysis of Puncher and Harrison (2012b). The sampling regime is summarised in Table C.7.

C4.6 HTO and OBT

An uncertainty analysis of doses resulting from ingestion of HTO and OBT (as dietary OBT) by adult members of the public has been performed by Harrison et al (2002). That study considered uncertainties in the values of biokinetic parameters in the ICRP Publication 56 models for HTO and OBT (ICRP 1989) and the effect of uncertainty in relative biological effectiveness (RBE) of tritium beta emissions. The sampling regime derived by Harrison et al (2002) was adapted in this study to calculate uncertainties on effective dose per unit ingestion for 1 year olds, 10 year old and adult members of the public. It was assumed that ingested OBT represented tritium associated with typical dietary intakes (i.e bound to carbohydrate, fat and protein). The sampling regime used by Harrison et al (2002) to model uncertainty on the central value of the partition between tritium in body water (compartment T_1) and organically bound tritium (compartment T_2) was used in this study. However, the distribution representing loss of tritium from these compartments employed in that study represents more closely variability in these processes in the human population. Harrison et al (2002) assumed a lognormal distribution with median value of 10 d^{-1} and GSD of 1.4 (95% rang $5\text{-}20 \text{ d}^{-1}$), and median of 63 d^{-1} and GSD of 1.8 (95% range $20\text{-}200 \text{ d}^{-1}$), to represent variability in the rate of loss from compartments T_1 and T_2 respectively. It is likely that there are sufficient data available describing these processes in the human population to make it possible to use the two-stage Monte Carlo approach described by Puncher and Harrison (2012a) to distinguish the effects of uncertainty and variability on estimating uncertainty on the population mean effective dose per unit intake. However, because of the computational burden entailed by implementing this approach, the present study approximated it by sampling the rate constants representing loss from compartments T_1 and T_2 from distributions that represent uncertainty on the population mean values, and then propagating these directly into the distribution of dose. This was implemented by multiplying the rate constants describing the loss of activity from T_1 and T_2 , by random variables sampled from lognormal distributions with median values of unity and GSDs of 1.2 and 1.6, respectively. This gives 95% ranges for the half life for the loss of activity from T_1 and T_2 of 7-14 days, and 16-102. These ranges were judged to give a reasonable coverage of the likely mean values based on the values collated by Harrison et al (2002); furthermore, the distribution assumed for T_2 covers the range of half-times of the long term pools of organically bound tritium in the body determined by Richardson and Dunford (2003a): 17 days (fat) and 62 days (protein). It was judged that using a one stage rather than two stage sampling regime introduced a small but insignificant bias and/or underestimation of the range of the estimate of effective dose per unit intake. However, the stated ranges probably slightly overestimate the uncertainty on the population mean values of these parameters and this compensates for any biasing, and also allows for further uncertainty in effective dose per unit intake arising from heterogeneity in the distribution of tritium in the body, noted by Harrison et al (2002). The sampling regime for HTO and OBT is summarised in Table C.9. Unlike the study of Harrison et al (2001), the present analysis did not consider uncertainty on the propensity of tritium beta emissions in causing radiogenic cancer

C4.7 Iodine

A number of studies have been published describing the application of parameter uncertainty analysis to calculate uncertainties on doses resulting from ingestion and inhalation of iodine compounds. These have been reviewed by Puncher and Harrison (2012a). The effective dose per unit ingestion of ^{131}I is determined by the amount of radioiodine taken up by the thyroid; doses from the much longer lived isotope, ^{129}I are also determined by the rate of removal of iodine from the thyroid. As noted earlier, the fraction of iodine taken up by the thyroid in 24 hours (the U_{24} value) is closely related to the amount of stable iodine present in the diet. Published uncertainty studies centre their distributions of thyroid uptake on a value of 25-30%. The ICRP Publication 56 model for iodine, which was used to calculate the dose coefficients for members of the public in ICRP Publication 72 (ICRP 1995a) predicts a U_{24} value for ^{129}I of 28%. These values accord with a population, such as that of the United States, with a relatively high dietary intake of stable iodine. However, a recent study by Vanderpump et al (2011) of daily urinary excretion of iodine by UK schoolgirls aged between 14-15 years suggests that the UK population may suffer from a mild iodine deficiency and thus UK residents would be expected to exhibit a higher U_{24} value and hence higher thyroid dose compared with a similar US population. From the median urinary concentration of stable iodine determined by Vanderpump et al (2011) and using empirical relationships between U_{24} and urinary excretion of stable iodine described by Stanbury et al (1954) and Leggett (2010), it can be inferred that the mean U_{24} value for a UK population is more likely to be around 40%, although there is significant uncertainty on this value. This estimate is consistent with a value of 44% determined for a Glasgow population in a study by Wayne et al (1964) and the relationship between U_{24} and daily urinary excretion of stable iodine observed in studies collated by Leggett (2010). For the present study, it was assumed that uncertainty on U_{24} for a UK population can be reasonably represented by multiplying the rate constant from blood to thyroid in the ICRP Publication 56 model for iodine by a random variable sampled from a lognormal distribution with median value of 2 and GSD of 1.4. This regime gave a mean value for U_{24} of 42% and a 95% range of 27-61% for ingested ^{129}I in Monte Carlo simulations; the simulated predictions followed a normal distribution. The value of 27% probably represents a conservative lower bound (a value more consistent with a population with optimum levels of dietary iodine, ~30%) and 61% a reasonable upper bound in the absence of more specific data. To model uncertainty on the rate of loss of radioiodine from the thyroid, it was assumed that this process represents secretion of thyroid hormone. Reference values for the rate of secretion collated by Leggett (2010) suggest a central range of 55-85 $\mu\text{g}/\text{d}$ for adults. In this study, uncertainty on the rate of removal of iodine from the thyroid was modelled by multiplying the reference rate of loss of iodine from the thyroid by a random variable sampled from a lognormal distribution with a median value of unity and GSD of 1.2. Assuming thyroidal iodine stores of 10 mg, this regime predicts a rate of secretion of between 60-124 $\mu\text{g d}^{-1}$ (the 95% range of simulated predictions).

To determine the effect of model uncertainty on doses from ingested or inhaled iodine, parameter uncertainty analyses were also performed using the revised systemic model for iodine published by Leggett (2010). Here, the sampling regime derived for the ICRP Publication 56 model was adapted for the new model as follows: the rate from compartment "blood 1" to compartment "thyroid 1" was multiplied by a random variable

sampled from a lognormal distribution with median value of 2 and GSD of 1.4. This regime gave a mean value for U_{24} of 45% and a 95% range of 29-62% for ingested ^{129}I . The rate from compartment “thyroid 2” to compartment “blood 2” was multiplied by a random variable sampled from a lognormal distribution with a median value of unity and GSD of 1.2 (the compartment names in quotations are those listed in Table 3 of Leggett 2010). Assuming thyroidal iodine stores of 10 mg, this regime predicts a rate of secretion of between 53-109 $\mu\text{g}/\text{d}$ (the 95% range of simulated predictions).

The sampling regimes for the systemic models for iodine are summarised in Table C.10.

Table C.1. Sensitivity analysis of effective dose per unit intake for ingestion and inhalation of radium by adults

Pathway/Parameter	Sensitivity Coefficient
Ingestion	
Fraction absorbed from the alimentary tract, f_1	0.98
Blood to upper large intestine	0.93
Trabecular bone surfaces (TBS) to blood	0.53
TBS to exchangeable bone volume	0.49
Non-exchangeable bone volume to blood	0.46
Blood to trabecular surfaces	0.20
Inhalation	
Fraction breathed through the nose, F_n	0.57
Slow dissolution rate, ss	0.51
Particle transport scaling factor, KPT	0.41

Table C.2. Probability distributions derived for the fraction absorbed from the alimentary tract to blood, f_1

Age group	Distribution	Median	GSD	
Uranium				
1 and 10 year old child	Lognormal	0.02	1.8	
Adult	Lognormal	0.02	1.6	
Plutonium and Americium				
All age groups	Lognormal	0.0005	1.73	
Organically bound tritium (OBT)				
All age groups	Uniform	0.9 ^a	1.0 ^b	
Caesium				
All age groups	Uniform	0.8 ^a	1.0 ^b	
Strontium				
1 and 10 year old child	Triangular	0.1	0.4	0.5
Adult	Triangular	0.1	0.3	0.4
Radium				
1 and 10 year old child	Triangular	0.05 ^c	0.3 ^d	0.4 ^e
Adult	Triangular	0.05 ^c	0.2 ^d	0.3 ^e
Iodine				
All age groups	Triangular	0.9 ^c	1.0 ^d	1.0 ^e

^aLower bound of uniform pdf; ^bupper bound of uniform pdf.

^cMinimum value of triangular pdf; ^dmode of triangular pdf; ^emaximum value of triangular pdf.

Table C.3. Probability distributions derived for HRTM parameter values

Parameter	Distribution	Median	GSD
Aerosol parameters			
AMAD	Lognormal	1.5	2.9
GSD	Lognormal	2.2	1.3
Breathing parameters			
F_n	Triangular	0.4 ^a	1 ^b 1 ^c
B	Lognormal	ICRP value ^d	1.3
f_R	Lognormal	ICRP value ^d	1.3
Particle transport			
ALV to bb1	Lognormal	0.0013	3.2
ALV to INT	Lognormal	0.001	4.5
INT to LN _{TH}	Lognormal	0.00003	3
Other rates scaled by factor K_{PT}	Lognormal	1	1.73
Dissolution parameters			
<i>Plutonium and americium</i>			
f_r	Lognormal	0.0026	3.1
s_r	Lognormal	1	4
s_s	Lognormal	0.000095	4.2
f_i	0.00001 (all ages)		
<i>Strontium and Caesium</i>			
f_r	Triangular	0 ^a	0.21 ^b 1 ^c
s_r	3 d ^{-1e}		
s_s	Uniform	0.001	0.005
f_i	$f_i=f_r$ (all ages)		
<i>Radium^g</i>			
f_r	Loguniform	0.001	0.1
s_s	Loguniform	0.00001	0.005
f_i	$f_i=0.3f_r$ (children)		
f_i	$f_i=0.2f_r$ (adults)		
<i>Uranium</i>			
f_r	Stepped uniform ^f	0.008-0.1	0.1-1
s_r	Lognormal	1	4
s_s	Lognormal	0.002	3.9
Vapour deposition			
<i>Iodine vapour (methyl iodide)</i>			
Regional lung deposition, V_{DP}	Normal	1 ^f	0.21 ^g

^aMinimum value of triangular pdf; ^bmode of triangular pdf; ^cmaximum value of triangular pdf.

^dThe ICRP reference values for a member of the public were varied together by multiplying each value by the same factor sampled from a lognormal distribution with median of unity and geometric standard deviation of 1.3.

^eValue for rapid dissolution rate for caesium aerosols was assumed to be 3 d⁻¹.

^fThe mean value of a normal pdf; ^gthe standard deviation of a normal pdf.

Table C.4. Lognormal distributions derived for the systemic model for uranium^a

Pathway	GSD
<i>Rates to skeleton from blood</i>	
BLOOD →CS/TS	1.3
<i>Rate from bone surfaces</i>	
CS/TS→other	3
<i>Rates from exchangeable bone volume</i>	
EXCH CV/TV→other	1.4
<i>Rates from non-exchangeable bone volume</i>	
NON-EXCH CV/TV→other	1.4
<i>Rate to soft tissues</i>	
BLOOD→ST2	1.73
<i>Blood to Liver</i>	
BLOOD →LIVER 1	1.4
<i>Blood to bladder</i>	
BLOOD →UBC	1.73

^a The rates for each of the given pathways were multiplied by a variable sampled from a lognormal distribution with a median of unity and the stated GSD (see main text); this enforces the correlations between rates discussed in the main text.

Key: CS/TS – Cortical/Trabecular Surface; EXCH CV/TV – exchangeable Cortical/Trabecular Volume; NON-EXCH CV/TV – non-exchangeable Cortical/Trabecular Volume; UBC – Urine Bladder Compartment; ST2 – long term soft tissue compartment

Table C.5. Lognormal distributions derived for the systemic models for strontium and radium^a

Pathway	GSD
<i>Rates to skeleton from blood</i>	
BLOOD →CS/TS	1.3
<i>Rate from bone surfaces</i>	
CS/TS→other	3
<i>Rates from exchangeable bone volume</i>	
EXCH CV/TV→other	1.4
<i>Rates from non-exchangeable bone volume</i>	
NON-EXCH CV/TV→other	1.4
<i>Rate to soft tissues</i>	
BLOOD→ST2	1.6
<i>Blood to upper large intestine</i>	
BLOOD →ULI	1.6
<i>Blood to bladder^b</i>	
BLOOD →UBC	1.6

^a The rates for each of the given pathways were multiplied by a variable sampled from a lognormal distribution with a median of unity and the stated GSD (see main text); this enforces the correlations between rates discussed in the main text.

^b Pathway not varied for radium.

Key: CS/TS – Cortical /Trabecular Surface; EXCH CV/TV – exchangeable Cortical/Trabecular Volume; NON-EXCH CV/TV – non-exchangeable Cortical/Trabecular Volume; ULI – Upper Large Intestine; ST2 – long term soft tissue compartment

Table C.6. Lognormal distributions derived for the systemic models for caesium

Pathway	GSD
ICRP Publication 56 model	
<i>Partition between compartments T_1 and T_2</i>	
A	1.6
$T_1 \rightarrow$ UBC/ULI	1.7
$T_2 \rightarrow$ UBC/ULI ^a	1.3
Leggett 2003 model	
<i>Rates to and from soft tissue^b</i>	
B	1.4

^a The median value for the half life representing loss from the long term compartment for adults was 100 days; for 1 and 10 year old children, the value was the ICRP Publication 56 value.

^b All rates to and from blood to soft tissue (liver, kidney, muscle, skeleton compartments, massive soft tissues) and from kidney to urine bladder were scaled by random variable *B*, sampled from a lognormal distribution with median of unity and GSD of 1.4

Key: A – partition coefficient from eqn. 1; T_1 – rate of loss from short term compartment T_1 in eqn. 1; T_2 – rate of loss from long term compartment T_2 in eqn. 1; UBC – Urine Bladder Compartment; ULI – Upper Large Intestine.

Table C.7. Lognormal distributions derived for the ICP Publication 67 systemic model for plutonium and americium^a

Pathway	GSD
<i>Rates to skeleton from blood</i>	
BLOOD \rightarrow CS/TS	1.3
<i>Rate from bone surfaces to bone volume</i>	
CS/TS \rightarrow CV/TV	1.73
<i>Rate from bone surfaces to marrow</i>	
CS/TS \rightarrow CM/TM	1.73
<i>Rates from liver to blood</i>	
LIVER \rightarrow BLOOD (Am)	1.73
LIVER2 \rightarrow BLOOD (Pu)	
<i>Rate to gonads</i>	
BLOOD \rightarrow OVARIES/TESTES	1.73
<i>Blood to bladder</i>	
BLOOD \rightarrow UBC	1.73
<i>Blood to bladder</i>	
BLOOD \rightarrow UBC	1.73

^a The rates for each of the given pathways were multiplied by a variable sampled from a lognormal distribution with a median of unity and the stated GSD (see main text).

Key: CS/TS – Cortical/Trabecular Surface; CV/TV – Cortical/Trabecular Volume; CM/TM – Cortical/Trabecular Marrow; UBC – Urine Bladder Compartment.

Table C.8. Lognormal distributions derived for the Leggett 2005 systemic model for plutonium^a

Pathway	GSD
<i>Rates to skeleton from blood</i>	
BLOOD1 →CS/TS	1.3
BLOOD1 →CV/TV	
<i>Rate from bone surfaces to bone volume</i>	
CS/TS→CV/TV	1.73
<i>Rate from bone surfaces to marrow</i>	
CS/TS→CM/TM	1.73
<i>Rates from liver to blood</i>	
LIVER1→BLOOD2	1.73
LIVER1→LIVER2	
<i>Rate to gonads</i>	
BLOOD1→OVARIES/TESTES	1.73
<i>Blood to bladder</i>	
BLOOD1 →UBC	1.73
BLOOD2 →UBC	

^a The rates for each of the given pathways were multiplied by a variable sampled from a lognormal distribution with a median of unity and the stated GSD (see main text).

Key: CS/TS – Cortical/Trabecular Surface; CV/TV – Cortical/Trabecular Volume; CM/TM – Cortical/Trabecular Marrow; UBC – Urine Bladder Compartment.

Table C.9. Lognormal distributions derived for the systemic model for tritiated water (HTO) and organically bound tritium (OBT)

Pathway	GSD
<i>Partition between T₁ and T₂</i>	
A (HTO)	Uniform(0.01, 0.1)
A (OBT)	Uniform(0.15, 0.75)
T1→Urine ^a	1.2
T2→Urine ^a	1.6

^aRates from compartments T₁ and T₂ in eqn. 2 multiplied by a random variable sampled from a lognormal distribution with median of unity and stated GSD.

Key: A – partition coefficient from eqn. 2; T₁ – rate of loss from body water compartment T₁ in eqn. 1; T₂ – rate of loss from organically bound tritium compartment T₂ in eqn. 1; UBC – Urine Bladder Compartment; ULI – Upper Large Intestine.

Table C.10. Lognormal distributions derived for the systemic models for iodine

Pathway	GSD
ICRP Publication 56 model	
BLOOD→THYROID ^a	1.4
THYROID→SOFT TISSUE	1.2
Leggett 2010 model	
BLOOD→THYROID 1 ^a	1.4
THYROID 2→BLOOD 2	1.2

^aThe rate from BLOOD/BLOOD 1 to THYROID/THYROID 1 was multiplied by a random variable with median value of 2; the remaining rates by a random variable with median value of unity.