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# Doses and risks from tritiated water and environmental organically bound tritium

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## Abstract

This short review provides an explanation of the calculation and use of the ICRP protection quantities, equivalent and effective dose, including the simplifications introduced by using radiation and tissue weighting factors. It discusses the dose coefficients ( $\text{Sv Bq}^{-1}$  intake) provided by ICRP for intakes of tritiated water (HTO) and organically bound tritium (OBT) and considers uncertainties in the human and animal data on which they are based, including information on the relative biological effectiveness (RBE) of tritium beta particles compared to gamma and x-rays. The review also addresses the specific issue of dose coefficients for ingestion of OBT in Cardiff Bay fish.

A distinction is drawn between the adequacy of the ICRP calculation of effective dose to a reference person for the purposes of planning and regulatory control, and the calculation of best estimates of dose and risk to individuals. ICRP will continue to use a radiation weighting factor of 1 for all low LET radiations in the calculation of effective dose, but specific RBE data should be used in risk estimates. Uncertainties in dose coefficients are small for HTO but greater for OBT. The generic consideration of OBT provided by ICRP may not be appropriate for specific organic forms such as OBT in fish.

## 1. Introduction

This short review considers the estimation of doses and risks from the ingestion of tritiated water (HTO) and environmental forms of organically bound tritium (OBT). The generally accepted authority on the biokinetics and dosimetry of radionuclides is the International Commission on Radiological Protection (ICRP). The biokinetic and dosimetric models developed by ICRP are used in the calculation of effective dose coefficients for use in the recommended protection system (ICRP 1991, 2007). However, the models are also used in the calculation of doses for other purposes, including epidemiological studies and calculations of probability of cancer causation (Harrison and Day 2008). It is important to distinguish, therefore, between the application of science in the calculation of effective dose coefficients,

which involves simplifying assumptions, and the use of best estimates of dose and risk, often with estimates of associated uncertainties.

Simplifications made in the calculation of effective dose include the use of radiation weighting factors to represent differences between radiation types in their ability to cause cancer per unit absorbed dose (Gy). Radiation weighting factors do not take account of all known differences between radiation types and energies for different tissues and cancer types (CERRIE 2004, Harrison and Day 2008). An advisory group of the HPA has recently issued a review on risks from tritium (HPA 2007) which includes the suggestion that ICRP should consider using a radiation weighting factor of 2 for tritium beta particles. However, as discussed by Cox *et al* (2008), the ICRP position is that a value of 1 will continue to be applied to all photons and electrons.

ICRP has provided dose coefficients for the ingestion of HTO and OBT for adults and children of different ages (ICRP 1989, 1993, 1996). Dose coefficients have also been published for exposures occurring *in utero* or due to breast-feeding, following intakes by the mother (ICRP 2001, 2004). In each case, the assumptions made regarding the biokinetics of OBT relate to unspecified organic components of food. Thus, the dose coefficients may not be applicable to specific forms of OBT. An example of some importance in the UK has been OBT in fish caught in the Cardiff Bay area. Studies using rats (Hodgson *et al* 2005) indicated that doses from this form of OBT might be somewhat greater than calculated using the ICRP dose coefficients (e.g.  $6 \times 10^{-11}$  Sv Bq<sup>-1</sup> for adults compared to the ICRP value of  $4.2 \times 10^{-11}$  Sv Bq<sup>-1</sup>). Recent volunteer studies, however, indicate that the lower dose coefficient for HTO ( $1.8 \times 10^{-11}$  Sv Bq<sup>-1</sup> for adults) might be more appropriate (Hunt *et al* 2009).

The following four sections of this review provide short discussions of:

- the use of the ICRP quantities, equivalent and effective dose;
- the assumptions made in calculating dose coefficients for HTO and OBT and associated uncertainties;
- the basis for the radiation weighting factor for tritium beta particles;
- the specific case of OBT in Cardiff Bay fish.

## 2. ICRP protection quantities

### 2.1. Biokinetic models

The first step in the calculation of doses from radionuclides taken into the body is the use of biokinetic models to represent the distribution and retention of elements and their radioisotopes in body organs and tissues. ICRP biokinetic models consider intakes by ingestion and inhalation by adults and children (ICRP 1979, 1980, 1981, 1989, 1993, 1994a, 1994b, 1995a, 1995b). Doses to the foetus following maternal intakes have also been calculated (ICRP 2001) and also doses to infants from radionuclides transferred to breast-milk (ICRP 2004). Models of the alimentary and respiratory tracts are used to define the movement of radionuclides within these systems, resulting in absorption to blood and/or loss from the body (ICRP 1979, 1994a, 2006). The behaviour of radionuclides absorbed to blood is described by element-specific systemic models (ICRP 1979, 1980, 1981, 1989, 1993, 1995a, 1995b). Systemic models range in complexity from very simple models that assume uniform whole-body distribution to multi-compartment recycling models that take account of movement within and between body organs and tissues. Thus, for example, the current models for tritium and isotopes of caesium consider uniform whole-body distribution with two components of retention while the models for strontium and plutonium are complex recycling models that represent uptake and retention in different skeletal tissues as well as other organs (ICRP 1989, 1993). As

discussed in section 3, ICRP (1989) models for tritium consider intakes as tritiated water (HTO) or organically bound tritium (OBT), with uniform whole-body retention of components representing HTO and OBT (half-times of retention of 10d and 40d, respectively, in adults; shorter in children).

The reliability of biokinetic models depends ultimately on the quality of the data on which they are based, including the availability of human data, but also on the realism of the model developed from these data (see section 3). For a number of elements and their radioisotopes, there are few or no human data for use in model development or validation, and reliance is placed on the results of animal experiments and chemical analogues. There are continuing efforts to provide improved models and over the next few years the ICRP's intention will be to publish new and updated models, and dose coefficients calculated using these models, to follow the new recommendations (ICRP 2007).

Biokinetic models for individual elements and their radioisotopes are used to calculate the number of radioactive decays (transformations) occurring within specific tissues, organs or body regions (termed 'source' regions) during a given period of time. The integration period used by ICRP in the calculation of committed doses is to age 70 y in all cases, applied to different ages of children and adults (age 20 y). The extent of protraction of dose over the integration period will depend on the decay characteristics of the radionuclide and the duration of its retention in body tissues.

## 2.2. Dosimetric models

Dosimetric models are used to calculate the deposition of energy in all important organs/tissues ('target' regions) for transformations occurring in each source region, taking account of the energies and yields of all emissions (Eckerman *et al* 1994). Absorbed dose in gray (Gy) can then be calculated, knowing the number of decays occurring in source regions and energy deposition in target regions.

Dose calculations rely on the use of reliable information on half-life, modes of decay, and the energies and yields of the various radiations emitted by nuclides and their progeny (Eckerman *et al* 1994, Endo *et al* 2003). Because the radiation types differ in their ranges in tissues, it is particularly important to account for the fraction of the available decay energy dissipated by conversion electrons, Auger electrons, and characteristic x-rays. Current calculations rely on nuclear decay data provided in Publication 38 (ICRP 1983) but new calculations will use more extensive updated information made available as Publication 107 (ICRP 2008).

Anthropomorphic phantoms are used to describe geometric relationship between different organs and tissues in the body. There are two main types of phantom—mathematical phantoms that approximate the sizes and shapes of organs mathematically (Cristy and Eckerman 1987) and voxel phantoms that are based on imaging data for real individuals, obtained using computed tomography or magnetic resonance imaging (Zankl *et al* 2002, 2003, 2007). ICRP currently uses mathematical phantoms (Eckerman *et al* 1994, Stabin *et al* 1999, ICRP 2001) but these will be replaced by models based on voxelised images (Zankl *et al* 2003, 2007, Fill *et al* 2004).

Doses from 'cross-fire' radiation between source and target regions (organs and tissues) are important for penetrating photon radiation. For 'non-penetrating' alpha and beta particle radiations, energy will in most cases be largely deposited in the tissue in which the radionuclide is deposited. For all dose calculations, radionuclides are assumed to be uniformly distributed throughout source regions, but while these are generally whole organs (e.g. liver), they may be a thin layer within a tissue (e.g. bone surfaces). Similarly, target cells for induction of

cancer and hereditary effects are assumed to be uniformly distributed throughout target regions but these vary in size from whole organs to layers of cells. As a consequence, source and target considerations are important for alpha and electron emissions in the specific cases of doses within the respiratory and alimentary tracts and the skeleton. Thus, doses are calculated to target layers within bronchial and intestinal epithelia from radionuclides in transit in the airways and gut lumen (ICRP 1979, 1994a, 2006, Harrison *et al* 2005, Phipps *et al* 2007) and to the whole or peripheral red bone marrow from radionuclides on bone surfaces or in bone mineral (ICRP 1979). Electron cross-fire is also taken into account in calculating doses to foetal tissues (ICRP 2001). An important concern is whether these assumptions provide adequate assessments of dose and risk, particularly when considering the heterogeneous distribution of short-range charged particle emissions (e.g. alpha emitters, low energy beta emitters such as tritium, and Auger emitters) in relation to target cells and their nuclei.

### 2.3. Radiation weighting factors

The next stage in the ICRP methodology is the transition from absorbed dose, a scientific quantity given the special name, gray (Gy), to the ICRP protection quantity, equivalent dose, with the special name, sievert (Sv). The calculation of equivalent dose provides a method by which the individual radiation doses to a given tissue or organ, from various types of ionising radiation (alpha, beta, gamma and x-rays), can be summed in relation to the effect they produce and, specifically, in relation to cancer induction. To achieve this objective, major simplifications are made.

Different types of radiation are known to vary in their effectiveness in causing cancer (ICRP 1991, 2003, 2007, UNSCEAR 2000). Currently, a simple one-dimensional indicator of ionisation track structure, namely the linear energy transfer or LET, is used to inform judgements on biological effects (ICRP 1991, 2003, UNSCEAR 2000). Clustered DNA damage, together with the degree of complexity of the damage, has been shown to increase with LET (Nikjoo *et al* 2002, ICRP 2003). The two broad categories of radiation that require consideration in the context of internal dosimetry are photons and charged particles, the latter including electrons and alpha particles. Photons and electrons (beta particles) are low LET radiations, alpha particles have high LET. However, very low energy electrons (e.g. Auger electrons, beta emission from tritium, electrons released by absorption of low energy x-rays) have higher LET values than higher energy beta particle emissions or electrons generated by conversion of gamma photons.

In practice, the assessment of the different effectiveness of different radiations relies on data on their relative biological effectiveness (RBE), defined as the ratio of the absorbed dose of a reference radiation to the absorbed dose of a test radiation required to produce the same level of effect. RBE is therefore an empirical quantity, which depends on the biological system, the observed end-point and the conditions of the experiment. Values are usually found to vary with dose and dose rate, increasing for high LET radiation to a maximum value at low dose and dose rate because of a curvilinear response at higher acute doses of the reference low LET radiation.  $RBE_{MAX}$  values are applicable to estimation of stochastic risk at low doses (ICRP 2003).

Low LET radiations show differences in RBE that reflect differences in their average ionisation density. Thus, for example, low energy beta emissions from tritium ( $^3\text{H}$ ) decay have been shown to have RBE values of up to between 2 and 3, compared to gamma rays, for *in vitro* end-points including cell killing, mutation and induction of chromosomal aberrations (Straume and Carsten 1993; see below). For photons, an increase in RBE with decreasing energy is supported by theoretical calculations (Nikjoo and Goodhead 1991) and experimental

observations (Hill 2004). Thus, compared to  $^{60}\text{Co}$  gamma rays, 29 kVp mammography x-rays have higher RBE values than 220 kVp x-rays by up to a factor of 4. Such RBE values have been observed for dicentric chromosome aberrations and cell transformation with mammalian cells *in vitro* (ICRP 2007).

Radiation weighting factors ( $w_R$ ) are chosen by ICRP as a simplified representation of the different effectiveness of radiations per unit absorbed dose in causing cancer. These simplifications are considered appropriate for the specified purposes of the protection quantities. Thus, despite differences in RBE between different low LET radiations and observations of different alpha particle RBE values for different end-points (UNSCEAR 2000, ICRP 2003, Harrison and Muirhead 2003), ICRP calculate equivalent dose using radiation weighting factors of 1 for all low LET radiations and 20 for alpha particles.

The equivalent dose,  $H_{T,R}$ , in tissue or organ  $T$  due to radiation  $R$ , is given by:

$$H_{T,R} = w_R D_{T,R}.$$

The total equivalent dose to an organ or tissue,  $H_T$ , is the sum of  $H_{T,R}$  over all radiation types:

$$H_T = \sum_R H_{T,R}.$$

#### 2.4. Tissue weighting factors

The purpose of the final stage in the ICRP methodology is to relate dose to risk in a simple manner, summing all radiation doses to all tissues in one risk-related protection quantity, the effective dose (Sv). To combine equivalent doses to different organs and tissues, tissue weighting factors ( $w_T$ ) are used to express the contribution of individual organs and tissues to overall detriment from cancer and hereditary effects, relating to whole-body radiation exposure.

The effective dose,  $E$ , is given by:

$$E = \sum_T w_T H_T.$$

Table 1 compares the  $w_T$  values used currently (ICRP 1991) and the values introduced in the new ICRP recommendations (ICRP 2007). The main source of data on cancer risks is the follow-up studies of the Japanese atomic bomb survivors. As well as longer follow-up, the new  $w_T$  values are based on cancer incidence rather than fatality data, adjusted for lethality and loss of quality of life. Weighting for hereditary effects is now based on estimates of disease in the first two generations rather than at theoretical equilibrium. The main changes in  $w_T$  values in the new recommendations are an increase for breast (from 0.05 to 0.12), a decrease for gonads (from 0.2 to 0.08) and inclusion of more organs and tissues in a larger 'Remainder' (from 0.05 to 0.12).

Tissue weighting factors are based on values of relative detriment, calculated separately for males and females and applying to populations of all ages. These relative detriment values and corresponding absolute detriment values are given in Annex A of the new recommendations (ICRP 2007). The overall detriment value for females is 40% greater than for males. The largest differences for individual organs are factors of 0.4, 0.5, 2.0 and 4.2 for females compared to males, for colon, liver, lung and thyroid, respectively. In addition, breast cancer accounts for about one-quarter of the total detriment in females. The male and female detriment and cancer incidence data tabulated by ICRP (2007) apply to populations of all ages. The BEIR VII Report (NAS/NRC 2006) provides estimates of life-time attributable risk for radiation exposure of males and females at different ages. These data show that risk estimates are generally about

**Table 1.** ICRP tissue weighting factors ( $w_T$ ).

Organ/tissue	ICRP (1991)	ICRP (2007)
Breast	0.05	0.12
Bone marrow	0.12	0.12
Colon	0.12	0.12
Lung	0.12	0.12
Remainder	0.05	0.12 <sup>a</sup>
Stomach	0.12	0.12
Gonads	0.20	0.08
Bladder	0.05	0.04
Liver	0.05	0.04
Oesophagus	0.05	0.04
Thyroid	0.05	0.04 <sup>c</sup>
Bone surfaces	0.01	0.01
Brain <sup>d</sup>	—	0.01
Salivary glands <sup>d</sup>	—	0.01
Skin	0.01	0.01

<sup>a</sup> The ICRP (2007) weighting factor of 0.12 for Remainder is apportioned equally between 13 organs/tissues in males and females: adrenals, extrathoracic tissue, gall bladder, heart, kidneys, lymphatic nodes, muscle, oral mucosa, pancreas, prostate ( $\sigma$ ), small intestine, spleen, thymus, uterus/cervix ( $\varphi$ ).

<sup>b</sup> The weighting factor for the thyroid was set at 0.04 to take account of evidence of a pronounced elevation of risk in childhood.

<sup>c</sup> Salivary glands and brain were given weighting factors of 0.01 because cancer risks, while not separately quantifiable, were judged to be greater than for other tissue in the Remainder.

double for irradiation in infancy compared with age 20 y, and about 5–6 times greater for thyroid cancer.

In applying the risk estimates derived from the A-bomb survivor data to cancer risks at low doses and dose rates, ICRP apply an empirical correction factor, the dose and dose rate effectiveness factor (DDREF), assuming a value of two for solid cancers (ICRP 1991, 2007). This assumption that risks per unit dose are lower at lower doses and doses rates is based largely on animal and *in vitro* data showing curvilinear dose–response relationships for exposure to acute low LET radiation. Judgements on an appropriate value for DDREF depend on the weight given to different sources of data. On the basis of the A-bomb survivor data, it is not possible to distinguish between a DDREF of 1 (no DDREF) or 2 for solid cancers (UNSCEAR 2000, Preston *et al* 2003). Experimental data generally show greater values and the ICRP (2007) judgement is that a value of 2 should continue to be applied. The BEIR VII Committee (NAS/NRC 2006) undertook probabilistic analyses of dose–response data and obtained a modal value for DDREF of 1.5 for solid cancers. For leukaemia, the A-bomb survivor data are consistent with the use of a linear–quadratic dose–response relationship and DDREF is not applied.

Having obtained risk estimates for exposures at low doses of a few tens of mGy and below, ICRP assume a linear relationship between dose and risk. This ‘linear non-threshold’ assumption (LNT) is considered to be the best approach on current evidence for radiation protection purposes (Preston *et al* 2003, NCRP 2001, ICRP 2006, 2007). Indeed, the LNT assumption is essential for the operation of the current protection system, allowing the addition of external and internal doses of different magnitudes, with different temporal and spatial patterns of delivery. Nevertheless, the LNT dose–response remains controversial, with arguments for supra-linear low dose responses and for thresholds and/or hormetic effects



(CERRIE 2004, French Academy of Sciences 2005, Feinendegen *et al* 2008). ICRP (2007) conclude that the true validity of the LNT model may prove to be beyond definitive resolution for the foreseeable future.

### 2.5. Use of equivalent and effective dose

The ICRP publishes dose coefficients ( $\text{Sv Bq}^{-1}$ ) for intakes of individual radionuclides, giving values of committed equivalent dose to individual organs and tissues, and committed effective dose (ICRP 1996, 2001). ICRP dose coefficients are calculated using defined biokinetic and dosimetric models, including reference anatomical data for the organs and tissues of the human body. They are calculated for reference adults, children of different ages and the foetus at different stages of development. They do not take account of individual characteristics. Radiation weighting factors are chosen as a simple representation of the different effectiveness of different radiations in causing stochastic effects at low doses and dose rates. They do not take account, for example, of observed differences between low LET radiations (e.g. photons of different energies), and of different alpha particle RBE values for different cancer types. A single set of tissue weighting factors is used to take account of the contribution of individual organs and tissues to overall detriment from cancer and hereditary effects, despite age- and gender-related differences. Doses to male and female adults will in future be calculated separately using new anatomical models but equivalent doses to males and females will be averaged before calculation of effective dose.

As discussed by Harrison and Day (2008), there is an apparent inconsistency between the increasing realism and complexity of biokinetic and dosimetric models and the continued use of a simple sets of radiation and tissue weighting factors. However, while the biokinetic and dosimetric models are primarily intended for use in the calculation of ICRP dose coefficients, they can be more widely applied. For example, they can be and are used to calculate absorbed doses to specific organs and tissues, both in the assessment of risks of stochastic effects and in the assessment of deterministic effects at higher doses. The models can also be used to provide dose estimates for epidemiological studies and in probability of causation calculations. The new generation of adjustable computational phantoms is ideally suited for these other applications. In contrast, radiation and tissue weighting factors are to be used solely in the calculation of the ICRP protection quantities, to provide a method for comparing all radiation exposures with dose limits and constraints. It would not be practicable to devise an internationally applicable system that would take account of recognised age-, sex- and population-related differences in risk factors and differences between radiation types. Increased complexity would create a false impression of the certainty with which radiation risks at low doses are understood. Central to the ICRP system is the optimisation of protection below constraints (ICRP 2007); constrained optimisation should ensure appropriate levels of protection, using the protection quantities with their inherent simplifications.

## 3. Dose coefficients for HTO and OBT

### 3.1. Current assumptions and uncertainties

The International Commission on Radiological Protection (ICRP) provides models and calculates dose coefficients for intakes of tritium as HTO or OBT (ICRP 1989, 1993, 2001). The models consider intakes of HTO and OBT by ingestion and inhalation by adults and children and doses to the foetus following intakes by the mother. The models make a number of simplifying assumptions. First, it is assumed that absorption to blood is complete and



**Table 2.** Ranges on values of biokinetic parameters and RBE derived by Harrison *et al* (2002), considering ingestion as either tritiated water (HTO) or organically bound tritium (OBT) (ICRP values in brackets).

Parameter	HTO	OBT
Absorption to blood	1 (1)	0.9–0.99 (1)
Fraction incorporated into OBT in body tissues	0.01–0.1 (0.03)	0.15–0.75 (0.5)
Half-time of retention of HTO component in adults, days	5–20 (10)	5–20 (10)
Half-time of retention of OBT component in adults, days	20–200 (40)	20–200 (40)
Relative concentration in foetus:mother	1.2–2 (1.4)	1.2–2 (1.4)
RBE	1–2.5	1–2.5

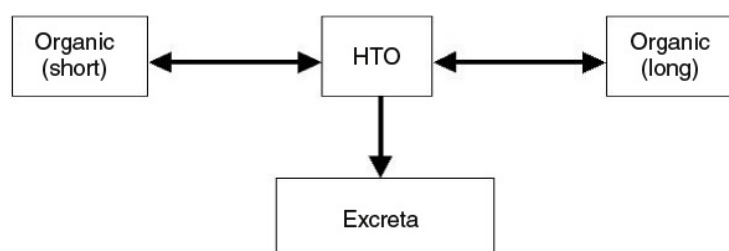
that tritium is subsequently distributed uniformly throughout body tissues. Second, retention in body tissues is represented by two components, the first corresponding to the turnover time of body water and the second to the turnover of carbon, with shorter retention times of both components in younger children. The first component corresponds to HTO and tritium exchangeably bound to organic molecules and essentially in equilibrium with body water, and the second component corresponds to non-exchangeably bound OBT. ICRP does not give dose coefficients for specific forms of OBT (e.g.  $^3\text{H}$ -DNA precursors)—dose coefficients for intakes of OBT are generic values for application to, for example, unspecified dietary intakes. For intakes of HTO, 97% is assumed to remain as HTO in the body and 3% is assumed to be incorporated into OBT in body tissues. The corresponding proportions after intake of OBT are assumed to be 50:50. For adults, the half-times of retention applying to the two fractions are 10 days for HTO and 40 days for OBT; half-times are progressively shorter at younger ages.

Harrison *et al* (2002) reviewed the experimental and human data on which the current ICRP dose coefficients for HTO and OBT are based and assessed the reliability of the dose coefficients in terms of uncertainties in central estimates for population groups. The analysis included uncertainties in the absorption of OBT to blood, incorporation of tritium into OBT in body tissues, retention times in tissues, transfer to the foetus and the relative biological effectiveness (RBE) of tritium beta emissions compared with gamma rays (table 2). Heterogeneity of dose within tissues and cells was also considered in the paper. The results of this analysis were 5%–95% uncertainty ranges on the central values of doses per unit intake for adults of about a factor of 3 for HTO and about 5 for OBT, with greater uncertainties for doses to children and the foetus (table 3). The central (50%) values from these distributions were about twice the corresponding ICRP values, largely because of the inclusion of a range of 1–2.5 for tritium's relative biological effectiveness (RBE). The consideration of OBT in this analysis applied to general dietary intakes and it was made clear that specific organic forms including DNA precursors should be considered on an individual basis.

Harrison *et al* (2002) did not recommend increases in the dose coefficients for HTO and OBT. The relationship between RBE and the radiation weighting factors used by ICRP is discussed below. It is also important to note that while such considerations of uncertainty may be instructive, ICRP regard their dose coefficients as not being subject to uncertainties. Strictly, the values given in table 3 are not effective dose (Sv) because they have not used defined ICRP models and weighting factors. Thus, ICRP dose coefficients are provided as point values for planning and demonstrating compliance with limits and constraints. Nevertheless, there may be circumstances in occupational and environmental exposures in which consideration of uncertainties might be valuable, including considerations of the optimisation of protection (Harrison and Day 2008).

**Table 3.** Probability distributions<sup>a</sup> on tritium doses from the ingestion of HTO or OBT by adults and for the foetus after maternal ingestion during pregnancy<sup>b</sup>, Sv Bq<sup>-1</sup> × 10<sup>11</sup>.

Age	Form	5%	50%	95%
Adult	HTO	2.1 <sup>c</sup>	3.9 <sup>d</sup>	6.6
	OBT	3.9	8.7 <sup>e</sup>	20
Foetus	HTO	3.7	7.6 <sup>f</sup>	14
	OBT	6.9	17 <sup>g</sup>	40

<sup>a</sup> Distributions on mean values for population groups.<sup>b</sup> Intake during pregnancy at 10 weeks after conception.<sup>c</sup> Read as  $2.1 \times 10^{-11}$  Sv Bq<sup>-1</sup>. Note that the tabulated values should strictly be Gy Bq<sup>-1</sup> × RBE since equivalent and effective doses are calculated using  $w_R$  values to represent RBE.<sup>d</sup> ICRP values are 1.8; <sup>e</sup> 4.2; <sup>f</sup> 3.6; <sup>g</sup> 7.6.**Figure 1.** The proposed systemic model for tritiated water.

### 3.2. Possible changes to ICRP assumptions for occupational intakes of HTO

Human data for exposures of adults to HTO provide evidence for a long-term component of retention of OBT in tissues with a half-time of between 140 and 550 days (Harrison *et al* 2002, Taylor 2003). A number of studies have shown two components of OBT retention after intakes of HTO, although in each case the first component had a half-time of less than 40 days (average about 30 days). Thus, these data do not invalidate the use by ICRP (1993) of a single value of 40 days to calculate dose coefficients. However, specification of components of retention may be important in the interpretation of urine measurements in retrospective dose assessments. To develop a biokinetic model for HTO that can be used for both the calculation of dose coefficients and the interpretation of bioassay data, the available data were re-evaluated by Taylor (2003) and three components of retention were proposed, with half-times of 10 days (99.00%), 40 days (0.98%) and 350 days (0.02%). Taylor (2003) pointed out that the data used to derive this model show quite wide variations both in the observed biological half-times and in the proportion of the total <sup>3</sup>H entering the long-term OBT component.

A Task Group of ICRP Committee 2 is currently preparing a report on assessment of dose from occupational intakes of radionuclides. The current draft includes a model for the systemic retention of tritium after HTO intake, shown in figure 1, with rates fitted to conform to three components of retention as proposed by Taylor (2003). Dose coefficients for HTO inhalation or ingestion by adults, calculated on the basis of these assumptions, are not significantly different from current ICRP (1993) values.

## 4. RBE and $w_R$ for tritium beta particles

Straume and Carsten (1993) provided a thorough review of experimental data on the carcinogenic, genetic, developmental and reproductive effects of exposure to HTO and OBT

in animals and *in vitro* cell systems. An advisory group to the UK Health Protection Agency has provided further review, updating and, in some cases, reanalysing data (HPA 2007). The spectrum of observed effects is indistinguishable from the effects of whole-body external irradiation with x-rays or gamma rays. Based mainly on *in vitro* studies, there is evidence that the RBE of tritium beta irradiation is generally greater than that of gamma irradiation and similar to or greater than x-irradiation. Although the observed effects of tritium are very largely attributable to ionisation damage, the transmutation of tritium to helium also has the potential to cause damage to DNA. Rapid dissolution of carbon–helium bonds will leave reactive carbon ions that can damage DNA by causing single-strand breaks and interstrand cross-links. The observed effects of tritium will include any contribution from such transmutation damage. Considering all observed effects of HTO exposure, RBE values were in the range of 1–3.5. For comparisons with gamma rays, most values were from 1–3 while for x-rays most were from 1–2, with values of 1–1.5 predominating. These measured RBEs for tritium beta irradiation are reasonably consistent with estimates based on microdosimetric considerations (e.g. Bigildeev *et al* 1992, Morstin *et al* 1993, Moiseenko *et al* 1997). For the purposes of assessing risk at low chronic doses, studies of carcinogenesis are the most appropriate. These include studies of the acceleration of the appearance of mammary tumours in rats (Gragtmans *et al* 1984) and the induction of acute myeloid leukaemia in mice (Johnson *et al* 1995). Both these studies compared chronic exposure to HTO or x-rays (250 kVp) and gave RBE values of about 1. Other carcinogenesis studies gave similar RBE values (Yokoro *et al* 1986, Cahill *et al* 1975a, 1975b). *In vitro* studies of transformation in 10T1/2 cells gave RBE values of up to about 2 compared to gamma rays. On the basis of a detailed analysis of a restricted number of studies considered to provide data of greatest relevance to carcinogenesis, the HPA advisory group (AGIR 2007, Little and Lambert 2007) obtained aggregated RBE estimates of 2.2 (95% CI of 2.0–2.3) and 1.2 (95% CI of 1.0–1.4), compared to chronic exposures to gamma rays and x-rays, respectively. These estimates necessarily rely on *in vitro* data.

An issue addressed by the HPA advisory group (HPA 2007) in general terms is the relationship between RBE values and DDREF. Different experimental systems exhibit different DDREF values and higher observed values of tritium RBE are likely to correlate with higher values of DDREF for the reference radiation (x-rays or gamma rays). Since RBE values for different low LET radiations will be smaller and tend to a value of one at high dose and dose rate, and given that there is also evidence for a small DDREF for tritium, the use of a DDREF of two for cancer induction in humans might be taken to suggest maximum RBE values for tritium induced solid cancers of between 1 and 2. On this basis, a best estimate of cancer risk from tritium in humans would be between the high dose rate and DDREF-corrected estimates for gamma rays. All such estimates are, of course, subject to uncertainties.

RBE data are used by ICRP (1991, 2007), together with biophysical considerations, as the basis for specifying radiation weighting factors ( $w_R$ ) used in the calculation of equivalent and effective dose. A value of 1 is applied to all low LET radiations, including tritium beta emissions, despite observed RBE differences of factors of 3–4. The reasonable justification is that in general the greater values may not apply to cancer induction in humans (ICRP 2007) and undue precision may be implied by the use of different  $w_R$  values for different low LET radiations. A single value of 20 is used for alpha particles despite evidence of differences for different cancer types. For example, an alpha RBE of 1–2 is probably more appropriate for the calculation of dose to red marrow to estimate the risk of leukaemia (UNSCEAR 2000, Harrison and Muirhead 2003). However, there are apparent inconsistencies in that a factor of four difference in neutron RBEs as a function of neutron energy is taken into account by a complex relationship between  $w_R$  and energy (ICRP 1991, 2007) and a value of 2 is now recommended for protons. It might be argued that a consistent scheme would use  $w_R$  values

of 1 for low LET radiations and protons and 10 for alpha particles and the charged particle component of neutron irradiation. Nevertheless, it is the responsibility of ICRP to define the methodology of calculation of their protection quantities, including the choice of radiation and tissue weighting factors.

## 5. Doses from OBT in Cardiff Bay fish

The study described by Hodgson *et al* (2005) was prompted by observations of unexpectedly high levels of tritium in fish caught in Cardiff Bay, considered to be attributable to discharges of tritium from the GE Healthcare plc plant (formerly Amersham), including a variety of forms of OBT (Williams *et al* 2001, Lambert 2001). The retention of tritium in adult rats was determined after administration as either tritiated water (HTO) or dried flounder flesh containing OBT. Two components of retention were obtained in each case. The first component, attributable to tritium equilibrating with body water, had a half-time of retention of about 3 days in each case, and accounted for about 97% of the intake as HTO and about 70% after intake of OBT in flounder. Results were consistent with the rapid catabolism of a proportion of flounder OBT to HTO. The second component of retention, attributable to OBT in rat tissues, accounted for about 3% of tritium after intake as HTO and 30% after intake as flounder OBT; the half-times of retention were about 10 days and 25 days, respectively. The results obtained for HTO administration are consistent with published animal data and correlate with ICRP assumptions for adult man of half-times of 10 days for 97% behaving as HTO in body tissues and 40 days for 3% incorporated into OBT in body tissues. The results obtained for flounder OBT suggest that appropriate assumptions for retention in adult man might be 70% with a 10 day half-time and 30% with a 100 day half-time. These assumptions result in an ingestion dose coefficient of  $6 \times 10^{-11}$  Sv Bq<sup>-1</sup> compared with the ICRP value for OBT of  $4.2 \times 10^{-11}$  Sv Bq<sup>-1</sup>. Including a longer-term component of OBT retention of 350 days, as done by Taylor (2003) (see section 3), would increase the flounder OBT dose coefficient from 6.1 to  $6.3 \times 10^{-11}$  Sv Bq<sup>-1</sup>. Hodgson *et al* (2005) proposed that a dose coefficient of  $6 \times 10^{-11}$  Sv Bq<sup>-1</sup> should be used for adults in calculating doses from the consumption of seafood from Cardiff Bay, unless it had been demonstrated that a significant proportion of the intake was HTO. Specific dose coefficients were also proposed for children.

Hunt *et al* (2009) present results of a study in which five volunteers consumed sole from Cardiff Bay and excreta were collected over periods of up to 150 days. The results suggested retention with half-times ranging from 4–11 days, with no evidence of a significant contribution due to retention with a longer half-life. It is not clear from these data whether a small longer-term component could have remained undetected. In addition, a large proportion of the ingested activity was unaccounted for and attributed to losses in sweat and exhaled air. However, the data are consistent with rapid catabolism to HTO and suggest that the most appropriate ingestion dose coefficients for tritium in Cardiff Bay sole might be those for HTO ( $1.6 \times 10^{-11}$  Sv Bq<sup>-1</sup> for adults).

## 6. Discussion

It is important to recognise that the ICRP protection quantities, equivalent and effective dose, do not relate to individuals but to reference persons. They are calculated using reference models and defined radiation and tissue weighting factors. They provide a method for the summation of doses from external radiation and from radionuclides that may differ substantially in their organ distribution and time-course of dose delivery. The protection quantities are intended mainly for planning and testing compliance with dose limits and constraints, set to limit the occurrence of stochastic effects. While equivalent doses will in the future be calculated separately using

male and female phantoms, practical protection would not be improved by calculating effective dose separately for males and females and to do so might give a misleading impression of precision. Similar considerations apply to the possibility of more complex treatments of radiation weighting and sex- and age-specific tissue weighting. Effective dose is not intended to provide estimates of doses and risks to individuals; for such calculations, best estimates of organ doses, RBE values and sex- and age-specific risk factors would be used, and it is likely that consideration of uncertainties would be appropriate.

Although ICRP dose coefficients are specified as point values without uncertainties, the data on which they are based are subject to uncertainties and it is possible, therefore, to assess uncertainties in dose coefficients. Strictly, the results of such an analysis should not be referred to as effective dose since they involve deviation from defined ICRP model parameter values and weighting factors. Harrison *et al* (2002) published an uncertainty analysis of dose coefficients for HTO and OBT. For intakes of HTO, dose is dominated by the short-term component of retention, attributable to the distribution of HTO in body water, with small contributions from the incorporation of tritium into OBT in body tissues. Since good human data are available for the turnover of body water in adults, uncertainties in doses from this component are small. Considering central values for population groups, adult dose coefficients for tritium as HTO can be regarded as known with high confidence, to within a factor of two. Turnover of body water in children provides a reliable basis for the calculation of HTO doses and the water content of the foetus is also known with reasonable confidence. Greater uncertainties in dose coefficients for OBT arise from uncertainties in the fraction incorporated into OBT in body tissues and the associated retention times since these are based on animal data.

The uncertainty analysis of Harrison *et al* (2002) included consideration of a range in RBE of 1–2.5. The review of Straume and Carsten (1993) concluded that for all observed effects of HTO exposure, RBE values were in the range of 1–3.5. For comparisons with gamma rays, most values were from 1 to 3 while for x-rays most were from 1 to 2, with values of 1–1.5 predominating. Kocher *et al* (2005) expressed tritium RBE using a log-normal distribution with geometric mean 2.4 and geometric standard deviation 1.4, implying a 95% CI of 1.2–5.0. It was not clear how they estimated the parameters of this distribution. They considered that tritium RBE could be up to 5 or greater and that a reasonable upper bound cannot be determined with certainty. A recent review of tritium RBE studies by an advisory group to the UK HPA (HPA 2007, Little and Lambert 2007) concentrated on a critical analysis of a restricted number of studies considered to be most reliable. They obtained an aggregate RBE estimate of 2.2 (95% CI 2.0, 2.3) with respect to chronic gamma radiation and 1.2 (95% CI 1.0, 1.4) with respect to chronic x-irradiation. Since the RBE data were obtained using radiations delivered at higher doses and dose rates than those generally received by workers or the public, it was concluded that values appropriate to human risk assessment could be greater. However, the conclusions reached were necessarily based mainly on the results of *in vitro* studies which have shown higher values of RBE than the small number of *in vivo* studies.

The HPA advisory group report on risks from tritium reached a number of conclusions regarding RBE (HPA 2007). Consistent with other reviews, it was concluded that the available data show a range of RBE values, mostly around 1–2 compared to x-rays and 2–3 compared to gamma rays. Given the dependence of RBE values on the reference radiation, the group recommend the use of a standard high energy  $\gamma$ -ray source such as  $^{60}\text{Co}$  as a reference for RBE studies and that an RBE value of 2 should be used for tritium epidemiological studies and retrospective dose assessments. Further, the report suggests that ICRP should consider adopting a value of 2 for the tritium radiation weighting factor ( $w_R$ ).

A clear distinction should be drawn between scientific best estimates of dose and risk and the ICRP protection system with its inherent simplifications and assumptions. For risk

assessments, the available data on RBE values for tritium beta particles compared to gamma rays suggest that an RBE value of 2 might be appropriate for cancer induction at low doses. However, this conclusion relies on the results of *in vitro* cellular studies, with little direct information on *in vivo* carcinogenesis. Considerations of the relationship between RBE and DDREF can be interpreted to suggest a low value of tritium RBE for cancer induction in humans at low doses and dose rates. The ICRP protection system is scientifically based but makes a number of simplifying assumptions so that doses from different radiation types can be summed and compared with limits, constraints, and reference levels that relate to whole-body radiation exposure. These underlying simplifications include the use of a simple set of radiation and tissue weighting factors and the assumption of a linear, no-threshold dose-response relationship at low doses. Cox *et al* (2008) have clarified the ICRP position, explaining why a  $w_R$  of 1 will continue to be used for all low LET radiations in the calculation of the ICRP protection quantities. By definition, the results of calculations using alternative values should not be referred to as equivalent and effective dose.

While ICRP provides dose coefficients of OBT, these apply to generic environmental forms and may not be appropriate for specific forms. In the case of OBT in Cardiff Bay fish, rat data for OBT in flounder (Hodgson *et al* 2005) suggest higher dose coefficients ( $6 \times 10^{-11}$  Sv Bq $^{-1}$  for adults compared with the ICRP value of  $4.2 \times 10^{-11}$  Sv Bq $^{-1}$ ), while human data for OBT in sole (Hunt *et al* 2009) are consistent with rapid catabolism and the use of dose coefficients for HTO ( $1.8 \times 10^{-11}$  Sv Bq $^{-1}$  for adults). Although greater weight will always be given to human data, it is probably appropriate, in view of these disparate results, to take the cautious approach of continuing to use the upper value derived from the rat study (EA 2007).

Cox *et al* (2008) concluded that ICRP should perhaps consider providing further general guidance on approaches to dose and risk assessment in different situations where equivalent and effective dose and standard dose coefficients may not apply. Concerning tritium, they agreed with the HPA advisory group (HPA 2007) on the importance of pursuing epidemiological studies on workers exposed to tritium. While it may be that these studies will lack the power to provide unequivocal conclusions, it is important that the most is made of opportunities to obtain human data. There is also a need for further *in vivo* carcinogenesis data, obtained in carefully conducted studies that closely match chronic dose delivery from tritium and gamma rays.

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