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"Emerging Issues on Tritium and Low Energy Beta Emitters"

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Working Party on Research Implications on Health and Safety Standards of the Article 31 Group of experts

Directorate-General for Energy and Transport
Directorate H — Nuclear Energy
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FOREWORD

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Under the terms of the Treaty establishing the European Atomic Energy Community, the Community, amongst other things, establishes uniform safety standards to protect the health of workers and of the general public against the dangers arising from ionizing radiation. The standards are approved by the Council, on a proposal from the Commission, established taking into account the opinion of the Group of Experts referred to in Article 31 of the Treaty. The most recent version of such standards is contained in Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation.

The European Commission organises every year, in cooperation with the Group of Experts referred to in Article 31 of the Euratom Treaty, a Scientific Seminar on emerging issues in Radiation Protection – generally addressing new research findings with potential policy or regulatory implications. Leading scientists are invited to present the status of scientific knowledge on the selected topic. Based on the outcome of the Scientific Seminar, the Group of Experts referred to in Article 31 of the Euratom Treaty may recommend research, regulatory or legislative initiatives. The European Commission takes into account the conclusions of the Experts when setting up its radiation protection programme. The Experts' conclusions are valuable input to the process of reviewing and potentially revising European radiation protection legislation.

In 2007, the Scientific Seminar discussed "Emerging issues on tritium and low energy beta emitters". Renowned scientists reported on the relevance of the concept of dose for low energy beta emitters, on metabolism, radiobiology and epidemiology of tritium, on tritium in the environment: sources, measurements and transfer, and on tritium in fusion facilities.

The Group of Experts discussed this information and drew conclusions that are relevant for consideration by the European Commission and other international bodies.

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CONTENTS

1 INTRODUCTION

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Tritium occurs from both natural and manufactured processes. As an isotope of hydrogen, tritium has extremely small natural abundance relative to hydrogen and deuterium. Tritium is naturally produced in the atmosphere by the interaction of high-energy cosmic radiation with oxygen and nitrogen and by ternary fission in geological formations. Tritium in the upper atmosphere is oxidized to tritiated water (HTO) and mixes with the surface waters of the earth.
Since 1954 (UNSCEAR, 1982), a major portion of the global inventory of tritium has resulted from the release of large amounts of manufactured tritium into the environment from nuclear weapon testing, nuclear power production, and industrial, commercial, and research uses of tritiated compounds. Much of the tritium that remains in the environment from nuclear weapon testing exists as HTO.
With the elimination by the U.S. and the ex-USSR of atmospheric nuclear weapon tests in the early 1960's, the concentration of tritium in the environment has been decreasing.

1.1 Characteristics of tritium

Tritium is an isotope of the element hydrogen which is both naturally occurring and manufactured. Its half-life is 12.26 years, decaying to helium (^{3}He) while emitting a beta particle. The beta particles, while of low energy (18.6 keV maximum, 5.7 keV average), have enough energy to produce ionizations and excitations of molecules in their path. Tritium poses no external hazard since the beta particles released during tritium decay cannot penetrate the outer layer of dead skin cells due to their average range in tissue of less than 1 μm, and maximum range of only 6 μm (ICRP, 1983). Because of the low beta energy, dilution throughout all of the soft tissues, and elimination with an average biological half-life of around ten days in adults, tritium as HTO (tritiated water) has relatively low radiological toxicity when compared to other pure beta emitters, such as ^{32}P or ^{90}Sr, or to common beta-emitters, such as ^{131}I or ^{137}Cs (ICRP, 1979-82).

1.2 HTO metabolism

Although tritium is not considered as a particularly toxic radionuclide, it presents a concern since it can become part of the biologically necessary hydrogen pool. If released into the environment, the tritium poses a potential internal radiation hazard since compounds containing tritium undergo various chemical transformations resulting in forms which can enter in the body. The majority of tritium in the environment exists as HTO. Because of its mobility in the environment and its biological importance, water is one of the most important compounds of tritium. As HTO, tritium can enter in the body by inhalation, ingestion, or diffusion through the skin. Once inside the body, the HTO diffuses freely and rapidly across cellular membranes, equilibrating throughout the total body water pool. The uniform concentration of HTO will result in the radiation dose being uniformly distributed throughout the body.

On the other hand, the tritium from HTO may exchange with hydrogen atoms and thereby become incorporated into organic molecules. HTO is the primary chemical precursor for other chemical forms of tritium. Essentially any organic molecule can incorporate tritium in this manner. Each will have a specialized metabolism associated with it that may result in inhomogeneous distributions of tritium within the body, within individual organs, and even within individual cells.
In general, the more rapidly a molecule is turned over, the more tritium will be incorporated per unit of time and the more rapidly the tritium will be removed from the same molecule. For longer-lived molecules, such as the structural protein collagen, or the phospholipids of some nerve cells, fewer tritium atoms will be incorporated per unit of time and those that are incorporated will be retained for longer periods of time (NCRP, 1979).

1.3 OBT metabolism

Besides HT and HTO, other forms of tritium are also found in the workplace. These forms include, but are not limited to, tritiated gases such as methane, metal tritides, tritiated pump oil and solvents, luminous plastic compounds containing tritium, and hundreds of soluble organic tritium-labelled compounds which are used in biomedical research. The chemical properties as well as the metabolism and retention of these tritiated compounds are largely unknown; however, some general assumptions based on known metabolic parameters for other tritiated compounds can be used to give a rough approximation of the metabolic patterns of these tritiated compounds (Taylor, 1990).
For radiation protection purposes, inhomogeneous distribution of tritium at the subcellular level as a result of exposure to HTO is generally not considered; however, localization of other tritium compounds occurs in certain cells or tissues and may cause biological effects that do not resemble those seen with HTO. An inhomogeneous concentration of tritium will generally result in doses to the involved cells that are larger than the average tissue dose, which may be negligible. As a result, the concept of average tissue dose for many tritiated compounds especially would be inappropriate and a poor estimate of the hazard associated from exposure to such compounds, especially if the exposure results in the tritium being incorporated into sensitive regions of the cells, such as DNA.
Little direct information is available on the metabolism of organically bound tritium (OBT) in humans. OBT can enter the body directly by ingestion of tritiated foodstuffs, by inhalation of volatile organic vapors or aerosols, or can be formed *in vivo* from tritium that is present in the general body pools after exposure to other tritium-containing compounds. Once in the body, OBT can be incorporated into a variety of biochemical compounds such as amino-acids, sugars, proteins, and other structural materials (Saito and Ishida, 1989). Organic molecules, in general, have slower turnovers and are retained in the body for longer periods of time than water. Tritium found in non-exchangeable organic binding sites (i.e., tritium bound to carbon atoms) will contribute an increased average soft-tissue dose over that seen for HTO alone, primarily due to the longer retention times in the body (ranging from hours to years) and inhomogeneous distribution among the different organs (Taylor, 1990). Analysis of observations of tritium elimination following accidental inhalation or ingestion of tritium has shown that up to three exponential components have been used to describe the retention curves. Besides the HTO biological half-life, two additional exponential components with longer biological half-times were used with ranges of 21-76 days and 280-550 days. Tritiated organic compounds that are metabolic precursors usually distribute in soft tissue and only rarely concentrate in particular cell types (ICRP, 1979-1982). The exception to this might be tritiated thymidine which, if not catabolized, is thought to reach the nucleus of cells that are actively synthesizing DNA. In this case, distributional and transmutational effects may result and should be considered in dose estimations. Very little, if anything, is known about metabolic processes and retention of these compounds. OBT, resulting from intakes of non

biological organic compounds containing tritium, is not expected to enter normal OBT pathways and the methods for obtaining accurate estimates of radiation dose from exposures of this type are not available. As a first approximation, uniform tissue distribution has been assumed as for HTO; however, for compounds classes such as solvents, the dose to the liver may prove to be of more concern than that for soft tissue of the body (Hill, 1993).
Many questions remain unanswered regarding the metabolism of OBT. While studies have shown that the knowledge available can be used to reliably estimate the uncertainties related to radiation exposure from OBT in foodstuffs for adults, questions rise when exposure to OBT is considered before adulthood is reached. If OBT is incorporated into molecules with very slow metabolic turnovers very early in life, it is not known how long tritium bound in this manner will remain in the body. More studies are needed to clarify the many questions associated with doses from OBT.

1.4 References

International Commission on Radiation Protection. Radionuclide transformations: Energy and intensity of emissions. New York: Pergamon Press; ICRP Publication 38; 1983.

International Commission on Radiation Protection. Limits for intakes of radionuclides by workers. New York: Pergamon Press; ICRP Publication 30; 1979-1982.

National Council on Radiation Protection. Tritium and other radionuclide labelled organic compounds incorporated in genetic material. Washington, DC: NCRP Report No. 63, 1979.

Taylor, D.M.; Moroni, J.P.; Snihs, J.O.;Richmond, C.R. The metabolism of H-3 and 14-C with special reference to radiation protection. Radiat. Protect. Dosim. 30:87-93; 1990.

Saito, M.; Ishida, M.R. Dose-modification factor for accumulated dose to cell nucleus due to protein-bound H-3. Health Phys. 56:869-874; 1989.

Hill, R.L.; Johnson, J.R. Metabolism and dosimetry of tritium. Health Phys. 65:629-647, 1993.

2 THE RELEVANCE OF DOSE FOR LOW-ENERGY BETA EMITTERS

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2.1 Preface

This text is intended is to set the scene for the remainder of the seminar, when subsequent presentations will consider in detail: metabolism, radiobiology and epidemiology of tritium; tritium and the environment; tritium in fusion facilities; and discussion on policy implications and research needs. Illustrations relating to the text are available in Goodhead (2007).

2.2 Introductory comments on dose, radiation quality and RBE

In radiation biology, clinical radiology and radiation protection the absorbed dose, D, is the basic physical quantity and is used for all types of radiation and any radiation geometry (ICRP 2007). Absorbed dose is defined as the quotient of dε by dm, where dε is the mean energy imparted to matter of mass dm (ICRU 1998). The units are joules per kilogram, which for absorbed dose is given the special name gray (Gy). The definition of absorbed dose has the scientific rigour required for a physical quantity. Absorbed dose is derived from the mean value of the stochastic quantity of energy imparted, ε, and does not reflect the random fluctuations of the radiation interaction events in tissue. While it is defined at a point in matter, its value is obtained as an average over a mass element dm. Absorbed dose is a physically measurable quantity. In radiation protection dosimetry, absorbed dose is usually averaged over whole organs or major tissue compartments.

The definition of absorbed dose is independent of the type ('quality') of radiation. The absorbed dose is approximately proportional to the average density of ionizations in a given mass or volume of interest, independent of the quality of radiation. However, the distribution of the ionizations within the volume is very strongly dependent on radiation quality, particularly on microscopic subcellular scales, but also on cellular and larger scales when doses or dose-rates are in the low ranges of main relevance in radiation protection. Radiation track structure determined the microscopic properties of the radiation in terms of the individual radiation tracks and the temporal relationships between them.

For a given absorbed dose of radiation, the biological effectiveness depends on a variety of additional factors, including type of radiation, dose rate or other temporal variations, and the particular biological system, effect and level of effect of interest. Therefore, absorbed dose alone is inadequate for practical radiation protection.

For the present seminar we are particularly concerned with consequences of radiation quality of tritium (and other low-energy beta emitters) in comparison with reference radiations, which are usually orthovoltage X-rays (typically ~ 220 kV) or gamma-rays from ^{60}Co (or ^{137}Cs). A number of other special features of low-energy beta (i.e. electron) emitters are also of interest.

All ionizing radiations produce charged particles and it is the tracks of ionizations along the paths of these particles that are predominantly responsible for the biological effects of the radiation. Electron tracks such as from gamma-rays, X-rays and beta-emitters are generally

classified as of low linear energy transfer (LET) due to their low average ionization density and, from the nanometre scale upwards, they contrast strongly with high-LET tracks, such as from alpha particles (Goodhead 1994a). The low-LET electron tracks show features of sparse scattered ionizations, as well as generation of low-energy secondary (including higher-order) electrons with regions of considerably higher ionization density from micrometre down to nanometre dimensions. The average dose to a typical cell nucleus (of diameter about 8 μm) from a single track (primary electron and its secondaries) from a reference ^{60}Co exposure is ~ 1 mGy. Another notable feature of typical gamma- or X-ray exposures is that ~ ¼ to ⅓ of the dose deposited in the biological material is via low-energy secondary electrons (Burch 1957, Nikjoo and Goodhead 1991).

Relative biological effectiveness (RBE) of a test radiation is defined as the ratio of the absorbed dose of a reference radiation to the absorbed dose of the test radiation to produce the same specific biological effect under identical irradiation conditions. The value of this experimental quantity in general depends on many factors, as mentioned above. Therefore, for a given type of radiation there is a range of RBE values, even with respect to a single reference radiation. Since dose-responses may be non-linear and have dose-rate dependence, the RBEs are frequently found to be a function of dose and of dose rate. It is a working assumption of the approach of the International Commission on Radiological Protection (ICRP) that dose responses for risks to human health become linear at low doses and that the ratios at minimal dose, RBE_M, define the maximum RBE of relevance in radiation protection (ICRP 2003). A further working assumption is that, for low-LET radiations, the slope of the low-dose response can be approximated by dividing the high-dose response by a dose- and dose-rate-effectiveness factor (DDREF), currently specified by ICRP as value 2 (ICRP 1991).

When comparing different radiations, there is a quite general trend for RBE values for a given system to increase with increasing LET of the radiation, at least up to high LET values of 100-200 keV/μm (corresponding to low-energy alpha particles). Although LET alone is an insufficient parameter to describe fully the biologically-relevant properties of radiation quality stemming from their track structures, it can provide some general semi-quantitative expectations.

2.3 ICRP system for radiation protection

The ICRP has developed a recommended system for radiation protection that is widely applied throughout the world (ICRP 1991). The main recommendations have recently been reviewed and only slightly altered (ICRP 2007). In both the current and the new recommendations, absorbed dose is the fundamental physical quantity on which radiation protection is based, but it is modified by two weighting factors in order to obtain additive dosimetric quantities, deemed to be of relevance in limiting human health risk at low doses and at low dose rates. The parameter values of both these subjective factors are prescribed by the ICRP on the basis of their committee judgements to represent what they deem to be reasonable simplifications and approximations to the available scientific information on biological effectiveness with respect to human detriment and to maintain a practically manageable system of radiation protection.

To allow for the differences in the radiation quality effect of various radiations in causing stochastic effects in humans, the ICRP defines equivalent dose, HT, in an organ or tissue as:

$$H_T = {}_R\Sigma(w_R D_{T,R})$$

where w_R is the radiation weighting factor for radiation R and $D_{T,R}$ is the absorbed dose in the tissue, T, due to radiation of type R (ICRP 2007). The units are J/kg, and for equivalent dose they are given the special name sievert (Sv).

To allow for variations in radiation sensitivity of different organs and tissues to the induction of stochastic effects, the ICRP defines *effective dose*, E, as:

$$E = {}_T\Sigma(w_T).{}_R\Sigma(w_R D_{T,R})$$
$$= {}_T\Sigma(w_T H_T)$$

where w_T is the tissue weighting factor for tissue T and $\Sigma w_T=1$, summing over all relevant tissues of the body (ICRP 2007). The units are J/kg, and for effective dose they are also given the special name sievert (Sv).

Thus, the radiation protection quantities equivalent dose and effective dose are not physical quantities. They involve two weighting factor values that are prescribed from judgement from available experimental radiobiological data, theoretical considerations, epidemiological studies of cancer induction and mortality after radiation exposure, as well as genetic data.

Primary risk estimates, mainly of cancer induction, have been obtained from epidemiological studies of radiation-exposed populations, especially the A-bomb survivors, as well as some medically-exposed groups, and are applied by the ICRP to obtain nominal risk probability coefficients for cancer for individual organs and the whole body, as well as to prescribe values for the tissue weighting factor. Hence, nominal estimated risks from *external* radiation exposure can be obtained by multiplying assessed absorbed doses to the organs or tissues of the body by the corresponding radiation weighting factors for the components of the incident radiation field, to obtain the equivalent doses to the organs, and then by the nominal risk probability coefficients for the organs. Further, the effective dose can be obtained by summing after application of the tissue weighting factors.

It should be noted, however, that the ICRP's recommendations for radiation protection are formulated in terms of effective dose (and in some cases equivalent dose) and not in terms of nominal risks, even though estimated risks are fundamental to the deliberations on which dose limits have been set. Thus, equivalent dose and effective dose act as surrogates for risk in radiation protection. The system contains many approximations and simplifications. It is quite complex, and yet quite crude, in order to obtain a system suitable for world-wide practical application in the workplace and the environment, including the advantages of additivity between radiation types, body distribution, temporal distribution, etc.

Similar methodology is applied for *internal* exposures, but in this case the absorbed dose to the tissues needs to be estimated by application of biokinetic models, to follow the location and time course of the radionuclide intakes throughout the body, and dosimetric models, to obtain the consequent doses to the organs. Again, application of w_R and w_T values provides the equivalent doses and the effective dose. This procedure allows derivation of dose coefficients for individual radionuclides, with some generalization for modes of intake and physical or chemical form, in terms of effective dose (or equivalent dose) per unit intake. Additivity of effective dose (and of equivalent doses to individual tissues) between internal and external radiation is achieved and these quantities remain appropriate for radiation protection from both internal and external radiation.

As explained by the ICRP (2007), effective dose is the primary quantity for dose limitation and optimization in radiation protection, for prospective dose assessments and planning and for demonstration of compliance with dose limits. Where individual retrospective dose assessments have to be made, more detailed information on the radiation field and appropriate RBE values may need to be considered if relevant data are available. For retrospective assessment of doses in specified individuals that may substantially exceed dose limits, effective dose can provide an approximate first measure of the overall detriment. If radiation dose and risk need to be assessed in a more accurate way, further specific estimates of organ or tissue doses are necessary, especially if organ-specific risks for the specified individual are needed. Effective dose is a quantity developed for radiation protection that is not suitable for use in epidemiological studies of radiation risks. Epidemiological analyses should be based

whenever available on estimates of absorbed doses to tissues and organs, taking full account of the circumstances of exposure and the characteristics of the exposed populations. Similarly organ or tissue doses, not effective doses, are required for calculations of probability of causation of cancer in exposed individuals.

2.4 Some issues for this symposium

Relevant issues include

- The appropriateness of the ICRP specification of w_R = 1 for *all* photon and electron radiations, including for low-energy beta emitters.
- Under what circumstances should this value be used (as, for example, for prospective planning, routine compliance records in radiation protection when doses are well below dose limits, etc)?
- What values of RBE should be used for particular low-energy beta emitters when more accurate doses or risk estimates are required (as, for example in retrospective dose assessments, prospective assessments and planning if doses might be a approaching dose limits, epidemiology, compensation, litigation, etc)?
- What other factors, in addition to radiation quality, may require consideration for particular low-energy beta emitters (for example, non-uniformity of absorbed dose to target cells within a tissue or to critical sub-cellular components, etc)?
- To what extent are these other factors incorporated into experimental measurements of RBE of low-energy beta emitters?
- The appropriateness of the ICRP tissue weighting factor, w_T, being the same for all radiation types, including low-energy beta emitters?

In both their 1990 recommendations and the recent modifications, the ICRP prescribes values of radiation weighting factor ranging from unity, for low-LET radiations, to 20 for alpha particles and heavy nuclei (ICRP 1991, ICRP 2007). Of particular note is that the single value of w_R = 1 is prescribed for *all* photon and electron radiations, internal and external and independent of energy. This is an issue of particular relevance to low-energy beta emitters. It is also of strong relevance to Auger-electron emitters, which will not be considered in this presentation. Use of this single value implies that, within the broad approximations of the ICRP system, all photons and electrons have the *same* risk per unit dose, irrespective of whether 'dose' is expressed as physical absorbed dose or the risk-weighted quantities equivalent dose or effective dose. The same radiation protection dose limits apply to them all. More pertinently, the ICRP treats absorbed dose from low-energy beta emitters (of energies of a few keV in some cases) exactly as if from orthovoltage X-rays (a few hundred keV) or high-energy gamma rays (~ MeV). This is despite substantial differences in their radiation qualities, based on their various track structures.

2.5 Beta decay of radionuclides

When a radionuclide undergoes beta-decay it emits a negatively-charged electron (β^- decay) or of a positively-charge positron (β^+ decay). In both cases, the atomic number of the decaying radionuclide remains unchanged in the daughter nuclide, but the nuclear charge is increased by +1 (in β^- decay) or decreased by -1 (in β^+ decay). A massless (or very nearly massless), uncharged particle is also emitted in either decay, namely an anti-neutrino in β^- decay or a

neutrino in β^+ decay. These features are in accordance with the relevant particle conservation laws. The total radionuclide decay energy is shared between the electron (or positron) and its corresponding anti-neutrino (or neutrino), with a very small proportion going to the recoil nucleus, according to the laws of conservation of momentum. Because this is a three-body decay process, sharing of energy between the electron and anti-neutrino depends on their relative angles of emission, with the result that the frequency spectrum of beta-particle energies ranges from energies of zero up to the maximum ('end-point') decay energy available. Due to charge effects, the energy spectrum of electrons (β^- particles) is skewed towards lower energies by electron/nuclear coulomb attraction and that of positrons (β^+ particles) towards higher energies by coulomb repulsion. An example of β^- decay is the decay of tritium (^3H), in which the maximum emitted electron energy is18.6 keV, but the mean electron energy is 5.7 keV (that is, less than one-third of the maximum).

Inspection of tables of common radionuclides (e.g. IARC 2001) shows a wide variety of beta-emitters, with energies ranging from a few tens of kiloelectron volts up to a few million electron volts and half lives varying from seconds to thousands of years. A notable feature of the low-energy beta-emitters is the very short ranges of their electrons, even less than a micrometer in some cases, compared to several millimetres for high-energy beta-emitters.

Positron-emitters (β^+ decay) tend to be of high energy, so are of little relevance to this symposium

2.6 Unusual features of low-energy beta emitters

2.6.1 Increased average ionization density sub-cellular (and cellular) scales

The stopping power of electrons increases as their energy decreases. Consequently, low-energy electrons have higher average ionization densities than do high-energy electrons or than the *average* ionisation densities of the electrons set in motion by the interaction of orthovoltage X-rays or gamma rays. There is no single measure of ionization density that can fully represent the differences of relevance to biological consequences or human detriment. The ICRP (1991) uses unrestricted linear energy transfer LET (L_∞) (that is, stopping power) as its sole measure of ionization density, when required. Alternative measures include energy-restricted LET, L_Δ, which applies a cut-off energy, Δ, to energy imparted by the primary particle and may then consider secondary electrons of energy $>\Delta$ as separate parts of the track to derive an average restricted LET (ICRU 1970). Averaging may be on a simple frequency (i.e. track) basis to yield the 'track-average', L_T, or by weighting by energy (i.e. L) to yield the 'dose-average'. Use of the dose-average LET would be appropriate if the effect of interest were precisely proportional to the square of the LET. The track-average LET has more general applications, including for special cases where the effect of interest is either independent of LET (e.g. 'hits') or is proportional to the LET. LET is a particularly limited concept in the case of electron (or photon) radiations because of substantial scattering of the primary and subsequent electrons and the very great diversity of features within the track, including sparse isolated ionizations from high-energy electrons but also very large numbers of low-energy secondary (and higher-order) electrons that are produced in the full slowing-down spectrum in all cases.

A different measure of ionization density has been proposed as lineal energy, y (ICRU 1983). Lineal energy is the quotient of ε by l, where ε is the energy imparted to the matter in a volume by a single energy-deposition event and $\bar{l}$ is the mean chord length in that volume (ICRU 1983). The mean lineal energy is obtained by randomly sampling individual tracks in the radiation field, averaging either on a simple frequency basis (frequency-mean lineal energy) or on an energy-weighted basis (dose-mean lineal energy). By convention, the single-event distributions do not include values for y = 0, so the distributions and their averages are

independent of absorbed dose. Use of the dose-mean lineal energy would be appropriate if the effect of interest were precisely proportional to the square of the lineal energy. The frequency-mean lineal energy has more general applications, including for special cases where the effect of interest is either independent of lineal energy (e.g. 'hits') or is directly proportional to the lineal energy (Goodhead 1987). Values of lineal energy, and ratios of lineal energy for different radiations, depend substantially on the size (and to some extent the shape) of the volume chosen for the determination. An ICRU/ICRP task group (ICRU 1986) focussed on spherical volumes ('sensitive sites') of 1 μm diameter, but this choice is not justified in terms of current knowledge of basic biology, radiation biology, theoretical considerations or the observed effects of radiation on human health effects (Goodhead 1987, 2006). No specific site size(s) have been shown to be appropriate in general, but there are many indications that much smaller dimensions are of particular relevance to the biological effects of radiation (Goodhead 2006).

Comparisons can be made of the average ionization density of tritium beta particles and reference X- and gamma-rays, on the basis of LET, or of lineal energy in selected target site volumes. Suitable data sets are, for example, in ICRU (1970) for LET, and Braby and Ellet (1971) and Booz (1976)) for lineal energies in sites of diameters 0.5 to 5 μm and Morstin et al (1993) for sites of 100 and 10 nm.

As pointed out above none of these measures of LET or lineal energy are adequate to provide reliable *quantitative* predictions of biological effectiveness or RBE. However, there is a very clear general trend for any of the LET or lineal energy measures to increase substantially from ^{60}Co gamma rays to orthovoltage X-rays and then to tritium beta particles. For example, tritium beta-particles have a restricted LET_{100} value of 4.7 keV/μm, whereas for ^{60}Co it is only 0.22 keV/μm. Only for low-energy X-rays (40-65 kV) are the values somewhat similar to tritium beta-particles.

Such data lead to the simple general conclusion that whichever of these parameters are used, the average ionization density of tritium beta-particles is greater than of orthovoltage X-rays, which in turn is greater than that of high-energy gamma rays. In view of the general tendency for biological effectiveness of radiations to increase with increasing LET, there should be a clear expectation that exposure to tritium is likely to yield an RBE > 1 for biological effects at the cellular level, at low, and possibly also high, doses.

As another comparison, 10 MeV protons have an unrestricted LET of 4.7 keV/μm (ICRU 1993) and restricted L_{100} of about half this value. The ICRP (1991) prescribed the radiation weighting factor w_R = 5 for protons of all energies above 2 MeV, but the new recommendations reduce this to w_R = 2, largely on the bases that practical proton radiations are only from external sources, protons of 10 MeV or less have very low penetration into the body (<1.2 mm) and higher energy protons have RBEs ~ 2 (ICRP 2007). Thus the ICRP incorporates a two-fold increased risk from protons in their radiation protection system, but it does not do so for tritium beta particles despite the fact that the LET of the latter is substantially greater than that of the protons.

Typical ionization patterns that occur in the terminal regions of the tracks of low-energy electrons, irrespective of whether they are primary or secondary electrons, can be revealed and analysed by Monte-Carlo track structure simulations of full interaction-by-interaction histories of the tracks. High ionization densities are readily visible on a scale comparable to the dimensions of a DNA molecule and this leads to the clear expectation of potential efficiency of such electron tracks to produce DNA double-stand breaks, as well as more complex forms of clustered damage, from either direct ionizations in the DNA, or from attack by hydroxyl radicals produced in the water within a few nanometres of the DNA, or from combinations of

both these mechanisms (Goodhead 1994a). These expectations have been quantified by Monte-Carlo simulations of the tracks together with the ensuing local chemistry leading to DNA damage and high yields of double-strand breaks have been confirmed experimentally (Goodhead 2006).

2.6.2 Short ranges of electrons (beta particles)

The ranges of the electrons emitted by beta decay of tritium, in particular, are very short even on the scale of individual cells or cell nuclei. For tritium decay, the maximum-energy beta particle has a range of about 7 μm and the average-energy beta has range 0.56 μm. For comparison, typical cells (or cell nuclei) have diameters from about 7 to 30 μm (nuclei about 6 to 15 μm), the diameter of elementary chromatin fibre is about 30 nm and contains the DNA of diameter 2.4 nm. Therefore, the short ranges of tritium beta particles do not mask their high-ionization-density features in subcellular structures such as DNA or elementary chromatin fibre. In principle, the short ranges could limit the ability of single tracks (of prime relevance at low doses) to damage two distant targets within a cell or in adjacent cells, leading to reduction in biological effectiveness if the effect were dependent on such double damage. Perhaps more importantly, the limited ranges would readily lead to inhomogeneity of dose if the beta emitters are themselves non-homogeneously distributed on the scale of the ranges of the beta particles.

2.6.3 Non-uniformity of absorbed dose

Due to the short ranges of the electrons from low-energy beta emitters, inhomogenities of absorbed dose will arise on all scales similar to or greater than the ranges of the electrons in situations where the beta emitting radionuclides are themselves inhogeneously distributed on these scales. As can be seen from comparison of the ranges given above and the sizes of biological structures, such inhomogeneities can apply to organs and tissue compartments for all low-energy beta emitters, to individual cells for several emitters, to major cell compartments for a few emitters and to chromosomes, chromatin and DNA for the lowest-energy emitters, most notably tritium.

As an example, the occurrence of such inhogeneities may arise at the DNA level (and upwards) from intake of tritiated DNA precursors, such as tritiated thymidine. This may be expected to increase the absorbed dose to DNA and chromatin relative to the average absorbed dose to cells or the tissue, and possibly to increase absorbed dose to actively proliferating cells relative to the average. Working in the opposite direction, in adipose tissue there may be excess accumulation of organically-bound tritium, particularly in females (Melintescu et al 2007), but the absorbed dose to target cells for cancer may be substantially lower than that assessed from the overall tissue dose.

It has also been suggested that the average ionization density from individual beta decays in target sites containing bound tritium (e.g. OBT) may be greater if the tritium is uniformly distributed (e.g. tritiated water). Calculations of Chen (2006) indicate that the enhancement of dose-mean lineal energy may be by a factor ~ 1.7 over a wide range of site diameters (10 nm to 1 μm). This radiation quality effect would be in addition to any dose increase in absorbed dose due to the inhomogeneity.

2.6.4 Cell (or nucleus) hit frequencies per unit dose

Even on the scale of a cell, or a nucleus, in the body, the average energy deposited by a single tritium beta-decay is about 4-fold greater than the average energy deposited by a single track from ^{60}Co gamma-ray exposure. Hence, equal low absorbed doses from tritium and ^{60}Co gamma-rays would correspond to about 4-fold fewer cells (or nuclei) being hit by tritium decay than from the gamma-ray exposure. In the case of low dose-rate exposures the time intervals between hits would also be longer for tritium decay. That is, the 'hits' from tritium beta-particles are larger in magnitude but correspondingly fewer in number. In principle, these differences

could have implications for the levels of biological effect at low doses and/or low dose rates, especially if threshold or bystander phenomena are involved.

In quantitative terms, for uniform irradiation with tritium the average specific energy deposited by a single beta track in a spherical volume of diameter 12 μm (or of diameter 7 μm) is 1.3 mGy (4.6 mGy), as compared to 0.4 mGy (1.1 mGy) for the average specific energy from a single electron track from ^{60}Co gamma rays. The reciprocal values provide the corresponding frequencies of an electron 'hitting' the sphere, namely for tritium 0.8 mGy^{-1} for 12 μm spheres (0.2 mGy^{-1} for 7 μm spheres) and for ^{60}Co 2.5 mGy^{-1} (0.9 mGy^{-1}), respectively. These illustrative cases of spheres of diameter 7 and 12 μm cover the range of a variety of typical cell or nuclear sizes. The hit frequencies have been calculated from the frequency-mean specific energies of single deposition events of the radiations, where the *specific energy (imparted)* is the quotient of ε by m, ε being the energy imparted by a single deposition event of the radiation to the matter of mass m (ICRU1983, Goodhead 1987).

2.6.5 Nuclear transmutations

During beta decay, the radionuclide transmutes to become a different element, with different chemical properties. Hence any molecule of which the radionuclide forms part will undergo some change and this may have biological consequences. Some examples are H to He, C to N and S to Cl.

2.6.6 Isotopic mass differences

By definition the mass of a radionuclide is different from the mass of the stable isotope(s) of the same element. The ratios of these mass differences are usually small for radionuclides of large mass, and even for most relatively light radionuclides, such as ^{14}C (ratio 7/6 relative to stable ^{12}C). However, the ratios can be large for some small radionuclides. For tritium in particular, the ratio is 3 compared to stable ^{1}H, the abundant stable isotope, leading to significant physico-chemical differences, including in diffusion, boiling point and triple point and reaction and clearance rates. Of particular interest is preferential binding of tritiated water in the hydration shells of DNA and in other macromolecules, including proteins. This exceedingly tightly bound water has been called 'buried tritium' (Baumgartner and Donhaerl 2004). It has been suggested that this fraction of tritium in bio-macromolecules, such as native proteins, is in bridge positions where the exchange rates are reduced from microseconds to days, months or even years. It has been estimated that enrichment factors for most macromolecules may be 1.4-fold but may be 2-fold in DNA. This topic is discussed in the report of AGIR (2007). The EMRAS working group of the IAEA discusses the nature and definition of organically bound tritium and debates if buried tritium must be considered together with tritium bound to carbon (Melintescu et al 2007).

2.6.7 Molecular

Different chemical forms of beta emitters, in common with their stable isotopes, can influence uptake ratios, retention times and other biokinetic parameters. Notable forms for tritium include tritiated water (important because of the abundance of water in the body), and organically bound tritium (OBT) in both exchangeable and non-exchangeable forms, including via DNA precursors.

2.6.8 Positron annihilation (β^+ emitters)

The annihilation of a positron with its anti-particle (electron) produces two photons of > 0.5 MeV by conversion of their rest masses. This process adds considerably to the energy resulting from β^+ decay, and also greatly delocalizes the resulting energy deposition in tissue.

In any case, β^+ emitters tend to have large decay energies, so they are unlikely to be of interest to this symposium on low-energy beta emitters.

2.6.9 Concluding remarks on special features

Many of the above-listed special features of low-energy beta emitters are not included in conventional radiation protection dosimetry.

It is useful to consider the extent to which they may or may not be incorporated into experimental measurements of RBE or other ratios of biological effectiveness. This will often depend on the particular experimental systems and protocols and the methods used for analysis of the data. For example, a factor 2 was reported by Ueno et al (1989) for mutagenesis of the tritiated DNA precursor ^{3}HTdR compared to HTO, in experiments with cultured mammalian cells. However, this comparison was based on estimates of dose to cell nuclei and converts to a factor of 6 on the basis of average cell dose (CERRIE 2004). It is possible that the ratio would be even higher in a tissue if there were a low concentration of ^{3}HTdR in the intercellular volumes.

2.7 A few additional comments

2.7.1 Low-energy electrons

All forms of low-LET radiation produce an abundance of low-energy electrons throughout the irradiation volume. This statement applies to photons of all ionizing energies, as well as to other electron-producing radiations. The presented diagrams illustrate that a substantial proportion of the dose deposited by very high-energy X-rays (2 MeV), ^{60}Co gamma-rays, orthovoltage X-rays, or electron beams, is via low-energy secondary electrons similar to those generated as primary or secondary electrons in the beta decay of tritium. The presented plots are based mostly on calculations of Burch (1957), which have been found to agree closely with results from Monte-Carlo track structure simulations (Nikjoo and Goodhead 1991). In these calculations energy transfers of less than 100 eV are deemed to be deposited locally by the electron of specified energy. It is notable that even for Co gamma rays, some 34% of the dose is deposited by electrons of energies < 5 keV. The corresponding figure for the 220 kV X-rays considered is 38% and for tritium beta-decay 77%. Even when only lower energy electrons are included, the dose proportions are substantial and a similar sequence applies for the various radiations. For example, below 1 keV, Co gamma rays deposit 27% of their dose, 220 kV X-rays 33% and tritium beta-decay 42%.

The relatively high ionization densities of low-energy electrons make them particularly efficient at generating double-strand breaks (DSB) in DNA, as mentioned earlier. Quantitative simulations and experiments with very low-energy ultrasoft X-rays (of 0.3 to 5 keV) have led to the suggestion that most double-strand breaks produced by low-LET radiations are due to individual low-energy electron tracks coinciding in or very near the DNA. Thus, multiple ionizations occur in or very close to the DNA, enabling two (or more) closely correlated strand breaks to be created either directly or by diffusion of hydroxyl radicals produced in the water sufficiently close by (a few nanometres). In experiments with mammalian cells, it was found that the RBE for induction of DSB rises from 1.4, for ultrasoft X-rays of energy 4.5 keV, up to 2.7 at 0.28 keV, with respect to ^{60}Co gamma rays (deLara et al 2001). These ultrasoft X-rays have also been found to be similarly effective in producing a wide range of permanent biological effects in cells, including mutations, cell inactivation, cell transformation and chromosome aberrations (Goodhead 1990, 1994, Hill et al 2001). Consequently it has been suggested that it is the low-energy component in all low-LET irradiations that is predominantly responsible for producing DSB and many cellular consequences (Goodhead 2006, Hill 2004).

In addition to the increase in yields of DSB by lower-energy electrons, Monte-Carlo simulations have predicted an increase in the complexity of the DSB (and other forms of clustered damage). The presented table shows a summary of results from Nikjoo, Goodhead and co-workers for irradiation by electrons of different initial energies. It can be seen, for example, that 32% of the DSB from 1 keV primary electrons are expected to be complex by virtue of there being at least one additional associated break in the cluster, as compared to 20% from 100 keV primary electrons (Goodhead 2006). Associated damage to DNA bases can add further to the complexity of DSB.

On the basis of such considerations, it may be expected that tritium beta-decay will produce a somewhat greater yield of DSB than the same absorbed dose of ^{60}Co gamma rays and that there may be a somewhat higher proportion of complex DSB, both of which factors may lead to greater biological effectiveness.

2.7.2 Reference radiation

Assessment of the relative effectiveness of low-energy beta emitters for purposes of radiation protection or risk estimation is confused by the lack of a generally agreed standard reference radiation for the comparisons. Commonly used reference radiations are orthovoltage X-rays (~100-300 kV) or gamma-rays (from ^{60}Co 1.2-1.3 MeV; or ^{137}Cs 0.66 MeV), whereas the main epidemiological base for human risk is from the A-bomb survivors exposed mostly to higher-energy gamma rays of 2-5 MeV. It is commonly stated that the RBE of orthovoltage X-rays with respect to ^{60}Co gamma rays is about 2. However, scientific justification for this claim is inadequate. Most of the literature, even to recent times (e.g. ICRP 2003) refers to small amounts of experimental data as quoted by Bond et al (1978), which are of questionable relevance to human risk. In addition, reliance has been placed very heavily on extensive data on dicentric chromosome aberrations in human lymphocytes irradiated *in vitro* (ICRP 2003). The appropriateness of the latter may be questioned on biological grounds and also on the reliance of parameter fitting to simplistic equations in this particular biological system to obtain extrapolated values at low dose (Goodhead 2000). A number of theoretical biophysical arguments have been put forward to suggest theoretical verification of this value of 2 (for example, based on dose-mean LET or lineal energy), but these tend to be circular arguments in that they seek and justify properties that yield the 'expected' factor 2 (ICRU 1986, ICRP 2003).

Thus, it remains difficult to relate the effectiveness of low-energy beta emitters to a clear base line of particular relevance for human risk.

2.7.3 Auger emitters

When Auger electrons are emitted as a result of radionuclide decay, they mostly have low (and even very-low) energies and therefore share many of the above unusual features of low-energy beta emitters. In general, however, the situation is more complicated for Auger emitters because of the great variety of decay properties from different Auger-emitting radionuclides, ranging from some where only a single Auger electron is emitted as a minor dose component of the overall decay, to others where a very large cascade of Auger electrons (up to more than 20) is the dominant feature of the radionuclide decay (for example ^{125}I decay). In the latter case, features of very large local energy deposition, overlapping of electrons (right down to the DNA level) and non-uniformity of emitters and of dose (on all scales of relevance) are likely to be much more severe than for low-energy beta emitters; much larger RBEs may be expected in some situations and have been observed in radiobiological experiments. Auger emitters are widely used in nuclear medicine.

2.8 Conclusions and recommendations

- It can be concluded that there is a general expectation, from theoretical and experimental considerations, that low-energy beta emitters will have greater biological effectiveness per unit absorbed dose than standard reference radiations under most circumstances. The effectiveness is expected to be greatest for the lowest-energy beta emitters, most notable tritium.
- The likely magnitudes of the relative effectiveness need to be established and practical implications considered, especially for tritium because of its low energy and its abundance.
- Some special features of low-energy beta emitters may not be included in routine RBE experiments.
- There may be issues with use of standard tissue weighting factors for all low energy beta emitters (e.g. access to target cells, or excesses therein). This has not been discussed.

Recommendations include:

- The use of available information, both experimental and theoretical, to establish the likely effectiveness of low-energy beta emitters for human risk relative to reference radiations,
- Consideration be given to special cases of potential practical relevance (e.g. extreme inhomogeneities).
- Experimental determination of yields and complexities of DNA damage (particularly DSB) from tritium beta emitters, including when bound to cellular DNA, in comparison with reference radiations.
- Seek agreement on single reference radiation of practical convenience and relevance to established human risk.
- As a practical solution for many applications, consideration should be given to applying a raised RBE, say of value 2, to all low-energy *internal* emitters. This would include low-energy beta emitters, soft X-ray emissions associated with a variety of radionuclide decays (these produce low-energy electrons, but are usually only a very small component of the overall decay energy and consequent dose), and Auger emitters (although further increases in RBEs are likely to be appropriate for the larger Auger-cascade decays). This solution should be fairly straightforward to apply. It would not include *external* irradiations with soft X rays, even though these also deposit most of their dose via low-energy electrons. For such X rays other factors are also relevant including the very high attenuation of the X rays as they pass into the body.

2.9 References

AGIR (2007). *Review of the Risks from Tritium*. Report of the Independent Group on Ionising Radiation, Documents of the Health Protection Agency, UK, RCE-4

Baumgartner, F and Donhaerl, W (2007). Non-exchangeable organically bound tritium (OBT): its real nature. *Anal Bioanal Chem*, **379**, 204-209.

Bond, V.P., Meinhold, C.B. and Rossi, H.H. (1978). Low dose RBE and Q for x ray compared to gamma ray radiations. *Health Phys*, **34**, 433-438.

Booz, J. (1975). In *Fifth Symposium on Microdosimetry* (Eds Booz et al), Commission of the European Community, Report EUR5452, pp 311-344.

Braby, L. A. and Ellet, W. H. (1971). Oregon State University, Report AD731709.

Burch, P.R.J. (1957). Some physical aspects of relative biological effectiveness. Brit J Radiol, **30**, 524-529.

CERRIE (2004). Report of the Committee Examining Radiation Risks of Internal Emitters (CERRIE). www.cerrie.org, ISBN 0-85951-545-1.

Chen, J. (2006). Radiat Prot Dosim, **122**, 546-548.

deLara, C., Hill, M.A., Jenner, T.J., Papworth, D. and O'Neill, P. (2001). Dependence of the yields of DNA double strand breaks in Chinese hamster V79-4 cells on the photon energy of ultrasoft X-rays. *Radiat Res*, **155**, 440-448.

Goodhead, D.T. (1987), Relationship of microdosimetric techniques to applications in biological systems. In *The Dosimetry of Ionizing Radiation*, Vol II (Eds K R Kase et al), Academic Press, pp1-89.

Goodhead, D.T. and Nikjoo, H. (1990). Current status of ultrasoft X rays and track structure analysis as tools for testing and developing biophysical models of radiation action. *Radiat Prot Dosim*, **31**, 343-350.

Goodhead, D.T. (1994a). Initial events in the cellular effects of ionizing radiations: clustered damage in DNA. *Int J Radiat Biol*, **65**, 7-17.

Goodhead, D.T. (1994b). Soft X-ray radiobiology and synchrotron radiation. In: *Synchrotron Radiation in Biosciences* (eds B.Chance et al) Oxford University Press, pp683-705.

Goodhead, D.T. (2000). Clustered damage in DNA: Time to re-evaluate the paradigms of radiation protection? In *Radiation Research Volume 2, Proc Eleventh International, Congress of Radiation Research, Dublin 18-23 July 1999* (Eds M. Moriarty et al). Lawrence, Allen Press, pp342-347.

Goodhead, D.T. (2006). Energy deposition stochastics and track structure: what about the target? *Radiat Prot Dosim* **122,** 1-15.

Goodhead, D.T. (2007). The relevance of dose for low-energy beta emitters. In: EU Scientific Seminar 2007 Emerging issues on tritium and low energy beta emitters. http://ec.europa.eu/energy/nuclear/radioprotection/seminars_en.htm

Hill, M.A., Stevens, D.L., Townsend, K.S.M. and Goodhead, D.T. (2001). Comments on the recently reported low biological effectiveness of ultrasoft X-rays. *Radiat Res,* **155**, 503-510.

Hill, M.A. (2004). The variation in biological effectiveness of X-rays and gamma rays with energy. *Radiat Prot Dosim*, **112**, 471-478.

IARC (2001). *Ionizing Radiation, Part 2: Some Internally Deposited Radionuclides*, IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol 78, IARC Press.

ICRP (1991). *1990 Recommendations of the International Commission on Radiological Protection*, ICRP Publication 60, Annals of the ICRP, **21** (1-3), Pergamon Press.

ICRP (2003). *Relative Biological Effectiveness (RBE), Quality Factor (Q), and Radiation Weighting Factor (w_R).* ICRP Publication 92, Annals of the ICRP, **33** (4), Pergamon Press.

ICRP (2007). *Recommendations of the International Commission on Radiological Protection*, ICRP Publication 103, Annals of the ICRP, **37** (2-4), Elsevier.

ICRU (1983) *Microdosimetry,* Report 36, International Commission on Radiation Units and Measurements, Bethesda.

ICRU (1986) *The Quality Factor in Radiation Protection*, Report 40, International Commission on Radiation Units and Measurements, Bethesda.

ICRU (1993) *Stopping Power and Ranges for Protons and Alpha Particles*, Report 49, International Commission on Radiation Units and Measurements, Bethesda.

ICRU (1998). *Fundamental Quantities and Units for Ionizing Radiation*, Report 60, International Commission on Radiation Units and Measurements, Bethesda.

Morstin, K., Kopec, M., Olko, P., Schmitz, T. and Feinendegen, L.E. (1993). Microdosimetry of tritium. Health Phys, **65**, 648-656.

Melintescu, A., Galeriu, D. and Takeda, H. (2007), *Radiat Prot Dosim* , pp 1-5 (doi:10.1093/rpd/ncm267)

Nikjoo, H. and Goodhead, D.T. (1991). Track structure analysis illustrating the prominent role of low-energy electrons in radiobiological effects of low-LET radiations. Phys Med Biol, **36**, 229-238.

Ueno, A.M., Furuno-Fukushi, I. and Matsudaira, H. (1990). In: *Tritium Radiobiology and Health Physics* (ed S Osaka) Proc 3rd Japan-US Workshop, Nagoya University, Japan. IPPJ-REV-3. pp200-210.

3 DOSES AND RISKS FROM TRITIATED WATER (HTO) AND ENVIRONMENTAL ORGANICALLY-BOUND TRITIUM (OBT)

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3.1 Issues addressed

This paper covers four main issues:

- The use of the ICRP quantities, equivalent and effective dose
- Dose coefficients for HTO and OBT, data used in their calculation and associated uncertainties
- The relationship between RBE, DDREF and w_R
- The specific case of OBT in Cardiff Bay fish

The protection quantities introduced and used by the International Commission on Radiological Protection (ICRP 1991, 2007) are described in some detail to provide an understanding of their intended purpose. This is followed by a description of the ICRP dose coefficients (Sv Bq^{-1} intake) for the ingestion and inhalation of tritiated water (HTO) and organically-bound tritium (OBT) and consideration of uncertainties in the data used in their calculation.

Important issue with regard to dose and risk estimates for tritium are the relationship between RBE data, showing that tritium beta particles are generally more effective in inducing cancer related end-points than are gamma rays, and the use by ICRP of a radiation weighting factor of 1 for all low LET radiations. The relationship between RBE and DDREF (Dose and Dose Rate Effectiveness Factor) is also of relevance.

Lastly, doses from a specific form of OBT are examined, namely OBT in Cardiff Bay fish, resulting from discharges from a radiochemical production plant.

3.2 ICRP protection quantities

3.2.1 Introduction

The International Commission on Radiological Protection (ICRP 1997, 1991) introduced the protection quantities, equivalent and effective dose, to facilitate the comparison of doses with dose limits and constraints. These quantities provide a method for the summation of contributions to dose and risk from external sources and radionuclides incorporated into body tissues, for the limitation of stochastic effects – cancer and hereditary effects. However, equivalent and effective dose and committed doses are often referred to simply as dose and there has been widespread misunderstanding of their intended use. There are also differences between experts on their interpretation (Edwards 2003).

Concerns over the use of the ICRP protection quantities are exemplified by the conclusions of a UK government committee, set up to examine the adequacy of ICRP methodology as applied to internal emitters. The Committee Examining Radiation Risks from Internal Emitters (CERRIE 2004) considered basic data on biological effects and epidemiology as well as

ICRP approaches to the estimation and limitation of doses and risks. Most members of CERRIE agreed that ICRP makes appropriate use of current knowledge. However, two important conclusions were that ICRP should clarify and elaborate its advice on the use of the quantities, equivalent and effective dose, and that more attention should be paid to uncertainties in dose and risk estimates and their implications (CERRIE 2004).

In the ICRP scheme, doses are calculated separately for adults, children of different ages, and for *in utero* irradiation of the embryo and fetus (ICRP 1996, 2001). However, a single set of tissue weighting factors (w_T) is used to represent the contributions of doses to individual organs and tissues to the overall risk of cancer and hereditary effects (ICRP 1991, 2007). These w_T values are chosen as averaged and rounded values on the basis of age- and gender- specific risk data. Adult organ and tissue doses will soon be calculated separately for males and females using newly developed reference anatomical models (Fill *et al*. 2004, Zankl *et al*. 2003, ICRP 2007). The male and female organ and tissue doses will be averaged before applying w_T values in the calculation of effective dose. Age and gender differences in doses and risks require that a clear rationale be given for both male / female dose averaging and the use of a single set of w_T values.

3.2.2 Calculation of equivalent and effective dose

Absorbed, equivalent and effective dose

The ICRP publishes dose coefficients (Sv Bq^{-1}) for intakes of individual radionuclides, giving both equivalent doses to individual organs and tissues, and effective dose (ICRP 1996, 2001). The steps in their calculation can be summarized as follows:

- The use of biokinetic models to represent the distribution and retention of radionuclides in body organs and tissues and calculate the total number of disintegrations occurring in each “source region”.
- The use of dosimetric models to calculate absorbed doses, D (Gy), to each target organ or tissue from disintegrations occurring in each source region.
- The use of radiation weighting factors, w_R, to take account of the relative biological effectiveness of different radiation types, converting absorbed doses to equivalent doses (Sv). 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:

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

- The use of tissue weighting factors, w_T, to represent the contribution of individual organs and tissues to overall detriment from cancer induction and hereditary effects, summing weighted equivalent doses to give effective dose (Sv):

  $$E = \sum_T w_T H_T$$

  where H_T is the equivalent dose in tissue or organ, *T*, and w_T is the weighting factor for tissue *T*.

The use of effective dose allows the summation of doses from different radionuclides and from external sources and comparison with dose limits set on the basis of risk relating to

whole body radiation exposure. Equivalent and effective doses are commonly 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. The steps in the ICRP scheme are examined below.

Biokinetic and dosimetric models

ICRP biokinetic models consider intakes by ingestion and inhalation by adults and children (ICRP 1994, 1996). Doses to the fetus following maternal intakes have 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 has recently developed a new model of the alimentary tract, which includes gender-dependent transit times for adults (ICRP, 2006). The behaviour of radionuclides absorbed to blood is described by element-specific systemic models that range in complexity from very simple models that assume uniform whole-body distribution (e.g. hydrogen, caesium) to multi-compartment recycling models that take account of movement within and between body organs and tissues (e.g. strontium, lead, uranium, plutonium). The most complex models are those developed for the bone-seeking alkaline earth and actinide elements. The representation of physiological reality in these models includes movement between organs and tissues via the circulation. In addition, the recycling models were designed to fit excretion data and can be used for bioassay interpretation. Simpler models for other elements are less suitable for this purpose.

Dose calculations involve the use of nuclear decay data (Endo *et al*. 2003) and anthropomorphic phantoms that describe geometric relationship between different tissues and organs. There are two main types of phantom – mathematical phantoms that approximate the sizes and shapes of organs mathematically (Eckerman et al. 1994) and voxel phantoms that use data for real individuals obtained using computed tomography or magnetic resonance imaging. ICRP currently uses mathematical phantoms that have been developed for adults and children of different ages (Eckerman *et al*. 1994), and for the pregnant woman and fetus for each trimester of pregnancy (ICRP 2001). Voxel phantoms for a reference adult male and female are currently being developed for use by ICRP (Fill *et al*. 2004, Zankl *et al.* 2003), adjusting data from scanned images for consistency with ICRP reference data for the body mass and related characteristics of adult males and females (ICRP 2002). Voxel phantoms for children will also be developed.

Doses from “cross-fire” radiation between source and target 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. However, source and target considerations are taken into account for alpha and electron emissions in a number of important cases. These include:

- doses to target cells in the walls of the bronchiolar airways from radionuclides in the mucus layer within the airway;
- doses to target regions in the gut from radionuclides in the lumen (ICRP 2006);
- doses to cells adjacent to inner bone surfaces (taken to be a 10 μm layer) and all red marrow from radionuclides on bone surfaces and within bone mineral;
- cross-fire irradiation between fetal tissues for electron emissions (ICRP 2001).

For all dose calculations, radionuclides are assumed to be uniformly distributed throughout source regions, although these can be whole organs (e.g. liver) or a thin layer within a tissue (e.g. bone surfaces). Similarly, target cells are assumed to be uniformly distributed throughout target regions that vary in size from whole organs to layers of cells.

Dose coefficients for children tend to be greater than for adults, depending on the radionuclide. For example, for ingestion of plutonium-239, the dose coefficient for a three month old infant is 17 times greater than for adults, while for caesium-137 the difference is a factor of 1.5. Differences between female and male adults, calculated using new anatomical models, will generally be smaller than between children and adults.

Radiation weighting factors

Different types of radiation are known to vary in their effectiveness in causing biological effects including cancer (ICRP 1991, 2003). These differences can be related in principle to the three- dimensional structure of ionisation tracks produced by charged particles traversing tissue volumes of interest, containing sensitive cellular targets including chromosomal DNA. The linking of biological effects to track structure is one of the central research goals in the field of microdosimetry. Currently, a simple one-dimensional indicator of track structure, namely the linear energy transfer or LET, is used to inform judgements on biological effects (ICRP 1991, 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.

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. It is 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}H) decay have been shown to have RBE values of up to between 2 – 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}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).

Reviews of available human and animal data on RBE for alpha-emitting radionuclides, comparing with low LET reference radiations, indicate that RBE depends on the biological end-point under consideration (ICRP 2003, Harrison and Muirhead 2003). Human data that allow comment on alpha particle RBE values are consistent with values of around 10 – 20 for lung and liver cancer and lower values for bone cancer and leukaemia, although considerable uncertainty must attach to any numerical estimate of RBE from these data (Harrison and Muirhead 2003). However, there is evidence from animal and *in vitro* studies of RBE values for alpha emitters of around 10 or greater for some cancer-related effects, and low values of 1 – 2 for leukaemia (UNSCEAR 2000). Human and animal data for bone cancer induction by alpha-emitting radionuclides suggest that there may be a threshold dose for some tumour types (UNSCEAR 2000, Harrison and Muirhead 2003).

Despite differences in RBE between different low LET radiations and observations of different alpha particle RBE values for different end-points, ICRP calculate equivalent dose using radiation weighting factors of 1 for all low LET radiations and 20 for alpha particles.

Tissue weighting factors

Tissue weighting factors (w_T) express the contribution of individual organs and tissues to overall detriment from cancer and hereditary effects, relating to whole body radiation exposure. Table 1 shows the values to be introduced in the new ICRP recommendations (ICRP 2007). As for the current values (ICRP 1991), the main source of data on cancer risks is the follow-up studies of the Japanese atomic bomb survivors. 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 the health effects annex of the new recommendations. 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.

Table 1. Tissue Weighting Factors, w_T, in new ICRP (2007) recommendations

Organ/Tissue	Number	w_T	Total Contribution
Lung, Stomach, Colon, Bone marrow, Breast, Remainder[a]	6	0.12	0.72
Gonads[b]	1	0.08	0.08
Thyroid, Oesophagus, Bladder, Liver	4	0.04	0.16
Bone surface, Skin, Brain, Salivary glands	4	0.01	0.04

[a]The specified remainder tissues (14 in total, 13 in each gender) are: Adrenals, Extrathoracic tissue (ET), Gall bladder, Heart, Kidneys, Lymphatic nodes, Muscle, Oral mucosa, Pancreas, Prostate(♂), Small intestine (SI), Spleen, Thymus, Uterus/cervix (♀).
[b]The w_T for gonads is applied to the mean of the doses to testes and ovaries.

Table 2. Life-time attributable risk of specific cancers after irradiation at different ages. Number of cases per 106 exposed to a single dose of 10 mGy. Selected data from BEIR VII (NRC/NAS 2006).

Cancer site	Age at exposure, years					
	Males			Females		
	0	20	60	0	20	60
Breast	-	-	-	1171	429	31
Colon	336	173	94	220	114	62
Liver	61	30	14	28	14	7
Lung	314	149	89	733	346	201
Thyroid	115	21	0.3	634	113	1
Leukaemia	237	96	82	185	71	57
All cancers	2563	977	489	4777	1646	586

The male and female detriment and cancer incidence data tabulated by ICRP (2007) apply to populations of all ages and take account of age-specific data. The BEIR VII report (NAS/NRC 2006) gives estimates of life-time attributable risk for radiation exposure of males and females at different ages. Selected data from the report are shown in Table 2. In general, risk estimates are about double for irradiation in infancy compared with age 20y, and about 5 – 6 times greater for thyroid cancer. Risks of *in utero* irradiation were considered in the context of setting tissue weighting factors by Streffer (2005), with the conclusion that contributions to overall detriment could not be reliably quantified on the basis of current evidence.

3.2.3 Use of equivalent and effective dose

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 fetus 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 soon be calculated separately using new anatomical models but equivalent doses to males and females will be averaged before calculation of effective dose.

It is clear from the approaches taken to the calculation of equivalent and effective dose that these quantities are not individual specific but relate to reference persons, to reference workers and reference members of the public of different ages. Their purpose is mainly for testing compliance with dose limits and constraints, providing a practical method for the assessment of doses received from internal exposure to different radionuclides and from external exposures. They are used in this way for regulatory purposes worldwide. Practical protection would not be improved by calculating effective dose separately for males and females and to do so would imply a degree of precision in the calculations that may be misleading. Similarly, a more complex treatment of radiation weighting or of tissue weighting for different age groups would be inconsistent with the intended purpose of the protection quantities and may again imply greater precision than is justified.

3.3 Dose coefficients for HTO and OBT

3.3.1 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 fetus following intakes by the mother. The models make a number of simplifying assumptions. First, it is assumed that absorption to blood is complete and 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 do not give dose coefficients for specific forms of OBT (e.g. ^{3}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 fetus and the relative biological effectiveness (RBE) of tritium beta emissions compared with gamma rays (Table 3). Heterogeneity of dose within tissues and cells was also considered in the paper. The results of this analysis were 5% to 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 fetus (Table 4). The central (50%) values from these distributions were about twice the corresponding ICRP values, largely because of the inclusion of a range of 1 to 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.

Table 3. Ranges on biokinetic parameters and RBE, considering ingestion as either HTO or OBT (ICRP values).

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 fetus: mother	1.2 – 2 (1.4)	1.2 – 2 (1.4)
RBE	1 – 2.5	1 – 2.5

Table 4. Probability distributions[a] on tritium doses from the ingestion of HTO or OBT by adults and for the fetus after maternal ingestion during pregnancy[b], Sv Bq^{-1} x 10^{11}

Age	Form	5%	50%	95%
Adult	HTO	2.1[c]	3.9[d]	6.6
	OBT	3.9	8.7[e]	20
Fetus	HTO	3.7	7.6[f]	14
	OBT	6.9	17[g]	40

[a]Distributions on mean values for population groups
[b]Intake during pregnancy at 10 weeks after conception
[c]Read as 2.1 x 10^{-11} Sv Bq^{-1}. Note that the tabulated values should strictly be Gy Bq^{-1} x RBE since equivalent and effective doses are calculated using w_R values to represent RBE.
ICRP values are [d]1.8, [e]4.2, [f]3.6, [g]7.6.

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 4 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.

3.3.2 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). Thus, 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) so 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 is clearly of importance 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 ^{3}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.

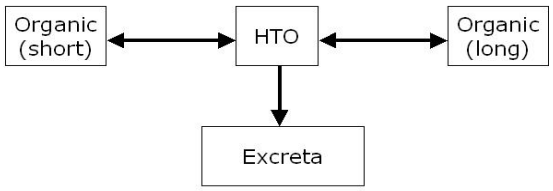

Figure 1. The general form of the proposed sytemic model for tritiated water.

3.4 The relationship between RBE, DDREF and w_R

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. The spectrum of observed effects is indistinguishable from the effects of whole body external irradiation with x rays or gamma rays. 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 ionization 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 a*l. 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 (Gragtams *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*. 1975). *In vitro* studies of transformation in 10T1/2 cells gave RBE values of up to about 2 compared to gamma rays.

These estimates of RBE were intended to apply to low doses and dose rates (Straume and Carsten 1993). The appropriate comparison is between tritium exposure and x-ray or gamma ray exposure over the same period of time, with the same temporal pattern of dose delivery. Some of the lower tritium RBE values reported in the literature are from studies in which the comparison is between protracted tritium exposure and acute exposure to the reference radiation at higher doses. RBE is generally dependent on DDREF – dose and dose rate effectiveness factor – which allows for the shape of the dose-response for low LET radiations, with maximum values of RBE applying at low doses and dose rates. Largely on the basis of animal data, ICRP use a DDREF of 2 in the calculation of risk factors for solid cancers at low doses and dose rates, based on the Japanese survivor data (ICRP, 1991, 2007). 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), 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 known 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 and 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 will be recommended for protons. A consistent scheme might 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.

3.5 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 *et al*. 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 tritium equilibrating with body water, had a halftime 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 x 10^{-11} Sv Bq^{-1} compared with the ICRP value of 4.2 x 10^{-11} Sv Bq^{-1}. Including a longer-term component of OBT retention of 350 days, as done by Taylor (2003) (see Section 2), would increase the flounder OBT dose coefficient from 6.1 to 6.3 x 10^{-11} Sv Bq^{-1}. Hodgson *et al*. (2005) proposed that a dose coefficient of 6 x 10^{-11} Sv Bq^{-1} should be used 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.

3.6 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 purpose of the protection quantities is mainly to test 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 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 fetus 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 final section of this paper (section 5) provides an analysis of the retention of tritium in rats after consumption of a specific form of OBT in Cardiff Bay fish. The study by Hodgson et al. (2005) was undertaken because of concerns that the ICRP generic dose coefficient for OBT might underestimate dose to fish consumers. The dose coefficient obtained from this study was about 50% greater than the ICRP value, suggesting that assessments using standard assumptions had been adequate.

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 - 3 while for x-rays most were from 1 - 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 to 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 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 2.19 (95% CI 2.04, 2.33) with respect to chronic gamma radiation and 1.17 (95% CI 0.96, 1.39) 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.

An advisory group to the UK HPA has produced a report on tritium (AGIR 2007) in which a number of conclusions are reached regarding RBE. Consistent with other reviews, it is concluded that the weight of evidence suggests that the RBE for tritium is 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 γ-ray source such as ^{60}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 distinction should be drawn between scientific best estimates of dose and risk and the ICRP protection system that has a number of inherent simplifications and assumptions. For risk assessments, the available data on RBE values of tritium irradiations compared to gamma ray irradiations 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 assumptions of a linear, no-threshold dose-response relationship. Since the EU Seminar, 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, of course, the results of calculations using alternative values should not be referred to as equivalent and effective dose.

3.7 References

AGIR (2007) Report of Independent Advisory Group on Ionising Radiation. Review of risks from tritium. Docs of HPA. RCE-4.

Bigildeev, E.A., Machalik, V. and Wilhelmovia, L. (1992) Theoretical estimation of quality factor for tritium. Health Phys. **63**, 462-463.

Cahill, D.F., Wright, J.F., Godbold, J.H., Ward, J.M., Laskey, J.W. and Tompkins, E.A. (1975) Neoplastic and life-span effects of chronic exposure to tritium. I. Effects on adult rats exposed during pregnancy. J. Natl. Cancer Inst. **55,** 371-374; II. Rats exposed *in utero*. J.Natl. Cancer Inst. **55**, 1165 – 1169.

CERRIE (2004) Report of the Committee Examining Radiation Risks of Internal Emitters. Avail. from the National Radiological Protection Board, Chilton. ISBN 0-85951-545-1.

Cox, R., Menzel, H.-G. and Preston, J. (2008) Internal dosimetry and tritium – the ICRP position. Invited Editorial. J. Radiol. Prot. 28, 131-135.

Diabaté, S. and Strack, S. (1993) Organically bound tritium. Health Phys. **65**, 698-712.

Edwards, A.A. (2003) The present ICRP protection quantities are seriously flawed. Opposing the proposition. Radiat. Prot. Dosim. **104**, 79-84.

Eckerman, K.F., Westfall, R.J., Ryman, J.C. and Cristy, M. (1994) Availability of nuclear decay data in electronic form, including beta spectra not previously published. Health Phys. **67**, 338-345.

Endo A, Yamaguchi Y and Eckerman KF (2003) Development and assessment of a new radioactive decay database use for dosimetry calculation. Radiat. Prot. Dosim. 105, 565-569.

Environment Agency (2001) Potential for bio-accumulation of organically-bound tritium in the environment. Review of Monitoring Data. National Compliance Assessment Service. Technical Report NCAS/TR/2000/026.

Fill, U.A., Zankl, M., Petoussi-Henss, N., Siebert, M. and Regulla, D. (2004) Adult female voxel models of different stature and photon conversion coefficients for radiation protection. Health Phys. 86, 253-272.

Goossens, L.H.J., Harrison, J.D., Kraan, B.C.P., Cooke, R.M., Harper, F.T. and Hora, S.C. (1997) Probabilistic accident consequence uncertainty analysis: uncertainty analysis for internal dosimetry. Vols. 1 and 2. Joint Report of the US Nuclear Regulatory Commission and the Commission of the European Communities. EUR 16773, Brussels.

Gragtmans, N.J., Myers, D.K., Johnson, J.R., Jones, A.R. and Johnson, L.D. (1984) Occurrence of mammary tumours in rats after exposure to tritium beta rays and 200 kVp X-rays. Radiat. Res. 99, 636-650.

Harrison, J.D., Khursheed, A. and Lambert, B. (2002) Uncertainties in dose coefficients for intakes of tritiated water and organically bound forms of tritium by members of the public. Radiat. Prot. Dosim. 98, 299-311.

Harrison, J.D. and Muirhead, C.R. (2003) Quantitative comparisons of cancer induction in humans by internally deposited radionuclides and external radiation. Int. J. Radiat. Biol. 79, 1-13.

Hodgson, A., Scott, J.E., Fell, T.P. and Harrison, J.D. (2005) Doses from the consumption of Cardiff Bay flounder containing organically-bound tritium. J Radiol. Prot. 25, 149-159.

Hill, M.A. (2004) The variation in biological effectiveness of x-rays and gamma rays with energy. Radiat. Prot. Dosim. 112, 471-481.

ICRP (1977) Recommendations of the International Commission on Radiological Protection. ICRP Publication 26. Pergamon Press, Oxford.

ICRP (1989) Age-dependent doses to Members of the Public from Intake of Radionuclides: Part 1. ICRP Publication 56, Ann. ICRP, 20(2). Elsevier Science Ltd, Oxford.

ICRP (1991) 1990 Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Annals of the ICRP 21 (1-3). Pergamon Press, Oxford.

ICRP (1993) 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). Elsevier Science Ltd, Oxford.

ICRP (1994) Dose Coefficients for Intake of Radionuclides by Workers. ICRP Publication 68. Annals of the ICRP 24(4). Elsevier Science Ltd., Oxford.

ICRP (1996) Age-dependent Doses to Members of the Public from Intake of Radionuclides Part 5: Compilation of Ingestion and Inhalation Dose Coefficients. ICRP Publication 72. Annals of the ICRP 26 (1).Elsevier Science Ltd, Oxford.

ICRP (2001) Doses to the Embryo and Fetus from Intakes of Radionuclides by the Mother. ICRP Publication 88. Ann. ICRP 31 (1-3). Elsevier Science Ltd, Oxford. Corrected version issued May 2002.

ICRP (2002) Basic anatomical and physiological data for use in radiological protection:

reference values. ICRP Publication 89. Annals of the ICRP 32 (3-4). Elsevier Science Ltd., Oxford.

ICRP (2003) Relative Biological Effectiveness (RBE), Quality Factor (Q), and Radiation Weighting Factor (w_R). ICRP Publication 92. Ann. ICRP 33 (4) Elsevier Sciences Ltd, Oxford.

ICRP (2004) Doses to infants from ingestion of radionuclides in mothers' milk. ICRP Publication 95. Annals of the ICRP 34 (3-4). Elsevier Sciences, Oxford.

ICRP (2006) Human Alimentary Tract Model for Radiological Protection. ICRP Publication 100. Annals of the ICRP 37 (2-4). Elsevier Science Ltd., Oxford.

ICRP (2007) 2007 Recommendations of the International Commission on Radiological Protection. (in press).

Johnson, J.R., Myers, D.K., Jackson, J.S., Dunford, D.W., Gragtams, N.J., Wyatt, H.M., Jones, A.R. and Percy, D.H. (1995) Relative biological effectiveness of tritium for induction of myeloid leukaemia. Radiat. Res. 144, 82-89.

Kocher DC, Apostoaei AI and Hoffman FO (2005). Radiation effectiveness factors for use in calculating probability of causation of radiogenic cancers. Health Phys, 89, 3–32.

Lambert, B. (2001) Invited editorial: Welsh tritium. J. Radiol. Prot. 21, 333-335.

Little MP and Lambert BE (2007). Systematic review of experimental studies on the relative biological effectiveness of tritium. Radiat. Environ. Biophys. [in press].

Morstin, K., Kopec, M., Olko, P., Schmitz, T. and Feinendegen, L.E. (1993) Microdosimetry of tritium. Health Phys. 65, 648-656.

NAS/NRC (2006) Health Risks from Exposure to Low Levels of Ionizing Radiation: BEIR VII Phase 2. Board on Radiation Effects Research.

NCRP (1979) Tritium and other radionuclide labelled organic compounds incorporated in genetic material. NCRP Report 63. Bethesda, Washington DC.

Nikjoo, H. and Goodhead, D.T. (1991) Track structure analysis illustrating the prominent role of low energy electrons in radiobiological effects of low-LET radiation. Phys. Med. Biol. 36, 229-238.

RIFE (2001) Radioactivity in Food and the Environment. RIFE-6. Food Standards Agency, Scottish Health Protection Agency.

Straume, T. and Carsten, A.L. (1993) Tritium radiobiology and relative biological effectiveness. Health Phys. 65, 657-672.

Streffer, C. (2005) Can tissue weighting factors be established for the embryo and fetus? Radiat. Prot. Dosim. 112, 519-523.

Taylor, D. M. (2003) A biokinetic model for predicting the retention of 3H in the human body after intakes of tritiated water. Radiat.Prot.Dosim. 105, 225-228.

Ueno, A.M., Furuno-Fukushi, I., Matsudaira, H. (1989) Cell killing and mutation to 6-thioguanine resistance after exposure to tritiated amino acids and tritiated thymidine in cultured mammalian cells. In: Tritium Radiobiology and Health Physics (Ed., S. Okada). Proc. 3rd Japanese-U.S. Workshop. Nagoya University, Japan. IPPJ-REV-3; pp.200-210.

UNSCEAR (1988) United Nations Scientific Committee on the Effects of Atomic Radiation. Sources, Effects and Risks of Ionizing Radiation. Report to the General Assembly, with Scientific Annexes. Annex. Early Effects in Man of High Doses of Radiation. United Nations, New York. Pp. 545-647.

UNSCEAR (2000) United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation. 2000 Report to the General Assembly, with Scientific Annexes. United Nations, New York.

Yokoro, K., Yamamoto, O., Seyama, T. and Kinomura, A. (1986) Acute and chronic effects of tritiated water in mice with special reference to its carcinogenicity." Radiat. Prot. Dosim. 16, 165-168.

Zankl, M., Petoussi-Henss, N., Fill, U. and Regulla, D. (2003) The application of voxel phantoms to the internal dosimetry of radionuclides. Radiat. Prot. Dosim. 105, 539-548.

4 THE RADIOBIOLOGY AND EPIDEMIOLOGY ASSOCIATED WITH EXPOSURE TO TRITIUM

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4.1 Reproductive Effects in the Female

Tritium taken into the body will be incorporated into most tissues and during pregnancy it will be taken up also by fetal oocyte DNA and other organic fractions during the five-month period from two months after fertilisation until seven months of gestation. This tritium will effectively irradiate the oocytes throughout the time before ovulation and fertilisation or before they are lost by atresia; this could in principle be over a time of 30+ years for tritium incorporated into DNA. These cells do not divide during this period and therefore, assuming no turnover of tritium, the half-time of elimination will be the physical half-life (12.26 years). Thus the dose to the nucleus will be greater than that which would be estimated using the ICRP half-time of 40 days. The dose to other cells in the foetus which have been labelled will decrease as the tritium incorporated is diluted by division.

4.1.1 Stability of DNA between oocyte formation and fertilisation

The argument that tritium incorporated into oocyte DNA during gestation remains there until fertilisation depends on there being no DNA turnover. If any DNA is replaced then the tritium incorporated therein will be lost. Damage to DNA occurs as a result of several spontaneous processes and the oocyte must either repair the damage or die. Early experiments showing DNA turnover in resting cells had their limitations but collectively they suggested that the phenomenon is widespread (see review by Pelc, 1968). For instance, in resting lymphocytes, Forell et al (1982) estimated around 2000 bases per hour were turned over. There is little quantitative evidence from oocytes but experiments with brief periods of incubation indicate that labelled thymidine is incorporated into the nuclei of resting, growing, and fully grown germinal-vesicle-stage mouse oocytes (Masui and Pedersen, 1975; Pedersen and Mangia, 1978). There is no reason to believe that the DNA of oocytes is immune to depurination, hydrolysis, and endogenous oxidative and methylating agents, all of which produce damage, and the repair of which involves DNA turnover. Repairable spontaneous depurination has been estimated to occur at a rate of 20,000 bases per day per mammalian cell, and over 50 years implies 7.3×10^6 purines are turned over in a human oocyte, about 2% of the genome. In addition to turnover due to DNA damage and repair there is likely to be a degree of turnover associated with postnatal synthesis of RNA in oocytes (NCRP, 1979) Of course, the frequency of such events in oocytes or how they are handled by the cell is unknown; their consequences might vary depending on their location within the oocyte genome. While the work described does not allow a quantitative estimate of DNA turnover, it serves to show that DNA in human oocytes should not be regarded as physically stable. Any calculations that assume complete persistence of tritium in oocyte DNA are thus likely to give an overestimate (possibly quite small) of the possible risk.

4.1.2 Doses from tritium in oocytes

In the ATSDR report (LLNL, 2002) on the implications of the tritium discharges from the Lawrence Livermore National Laboratory and the Savannah River Site in the USA, the effects on oocytes were assessed using an approach which involved the reasonable assumption that

the oocytes are labelled with tritium to the same concentration as the rest of the body. As an example of the implication of this exposure and uptake of tritium, a similar approach (calculating from first principles) is presented here. This is applied to people who live in the Cardiff Bay area of Wales where there is an accumulation of OBT in flounder (as a result of the discharges from a local radiopharmaceutical production facility).

Concentrations of OBT in a variety of fish species have been reported as ranging from 20–120 kBq/kg, but with most values between 20 and 50 kBq/kg (Williams et al, 2001).

The critical group consumption rate is taken to be 24 kg per year (EA et al, 2006), and assuming a concentration of 50 kBq/kg implies an intake of 1200 kBq of OBT per year.

Hodgson et al (2005) have developed a model for OBT ingested in Cardiff Bay flounders. Making the conservative assumption that the intake rate above has persisted for sufficiently long so that an equilibrium level has been reached in the critical group, then whole-body retention of 175 kBq using the Hodgson et al model can be calculated.

Assuming that this intake is distributed throughout all the maternal and fetal cells (i.e. about 10^{14} cells). The cell content is thus 1.75 nBq of tritium distributed throughout the cell constituents.

Around 5% (87.5 pBq) would be expected to have labelled the DNA. Assuming this DNA is not turned over until the time of possible fertilisation the number of disintegrations (N) in an oocyte in, say, 30 years will be as follows:

$$N = 87.5.\ 10^{-12} \int_0^T \exp(-\lambda t)dt$$

$$N = 87.5.\ 10^{-12} \times \lambda^{-1} [1 - \exp(-\lambda T)]$$

Where $\lambda = 0.693/T_{1/2}$ ($T_{1/2}$ = 12.26 years) and T is 30 years

$$N = 0.04 \text{ disintegrations per cell}$$

Thus, about 4% of the cells will experience a tritium disintegration in 30 years, 96% will not.

The authors of the ATSDR (LLNL) calculated that each disintegration generates a dose of 2.7 mGy on average in an assumed cell nucleus of 8 µm diameter and 270 pg mass. If adjustment is made for an RBE weighting factor of 2 for tritium, then those oocytes in which a disintegration has occurred will experience an adjusted dose of 2 x 2.7 = 5.4 mGy equivalent.

The ICRP value for the probability of severe hereditary effects is $5\ 10^{-6}$ per mGy based on extrapolation from male mouse data. For those oocytes that experience a tritium disintegration (equivalent to an adjusted dose of 5.4 mGy equivalent) accumulated between the formation of the oocyte and the laying down of an oocyte during a pregnancy 30 years later, the probability of a severe hereditary effect is thus likely to be around $2.5\ 10^{-5}$. However, since only 4% of oocytes will experience a tritium disintegration, the overall frequency will be around 10^{-6}.

For the critical group consuming Cardiff Bay flounder during pregnancy, with an assumed body content at equilibrium of 175 kBq based on cautious estimates of intake, it can be estimated that approximately 4% of oocytes could experience a tritium disintegration within the next 30 years.

The frequency of severe hereditary effects resulting from this would be expected to be around 10^{-6}. This may be compared with a spontaneous incidence of around 3 to 4%. Clearly, there are significant uncertainties in the calculation of the value of 10-6, nevertheless this is of little concern when the result is very small compared to the spontaneous incidence rate.

4.1.3 Response to DNA damage in the oocyte

It is worth considering whether some sort of amplification of radiation damage might occur during embryonic development, operating through, for example, persisting genomic instability, bystander effects, or clonal expansion. Alternatively, DNA damage responses such as cell cycle checkpoints and entry into apoptosis may be different in the rapidly expanding and differentiating cell populations of the embryo. An important property of genomic instability and the bystander effect is that their consequences are not limited to the cells experiencing the radiation dose but can (via signal transduction processes) occur in neighbouring cells or in the descendents of the irradiated cell. Effects of this nature have been reported to occur in irradiated male germ cells and may extend into the early cell divisions following fertilisation (for reviews of this area see Bridges, 2001, and Bouffler et al, 2006). Whether genomic instability and bystander effects occur in oocytes and throughout development is unknown, although there would appear to be considerable scope for the bystander effect. Only a very small proportion of oocytes experience a tritium decay and there will be long intervals between decays. It is unclear whether amplification effects such as those mentioned above could have a significant effect on pregnancy outcome other than the severe effects normally considered.

4.1.4 Conclusions

(a) Tritium incorporated during pregnancy into the DNA of fetal oocytes could in principle remain there until fertilisation decades later.

(b) DNA, however, is not stable since it is subject to turnover during DNA repair processes. Such evidence as is available suggests that even over decades, only a small proportion of the DNA will be turned over. A conservative assumption would be to neglect turnover for radiation protection and risk assessment purposes.

(c) For the critical group consuming Cardiff Bay flounder during pregnancy, it is estimated that approximately 4% of oocytes could experience a tritium disintegration within 30 years. The frequency of severe hereditary effects resulting from this would be expected to be around 10^{-6}. This may be compared with a spontaneous incidence of around 3 to 4%.

(d) Existing evidence does not enable account to be taken of any consequences there might be resulting from bystander effects or genomic instability phenomena in the developing embryo.

4.2 Radiobiological Effectiveness

Of prime importance for the assessment of risk posed by tritium is its relative biological effectiveness (RBE). Apart from considerations of track structure there are two further theoretical reasons why the RBE of tritium might be greater than one.:

(i) Transmutation of hydrogen to helium
(ii) Accumulation of tritium (particularly HTO) in the hydration shell of DNA.

(i) During the transformation to helium a maximum recoil energy of 3 eV is given to the helium nucleus – this has not been considered sufficient for bond breaking (Kacena 1967). The possibility that the actual transmutation which occurs when tritium decays could cause some separate effect has been considered, both experimentally and theoretically, by a number of authors (Feinendegen and Bond 1971, Feinendegen 1967, Krisch and Zelle

1969, Person et al 1976, Teebor et al 1984, Krasin et al 1976, Cleaver 1977, Tisljar-Lentulis et al 1983, Korolev 1985). The consensus from these studies was that very nearly all the effects seen could be accounted for on the basis of the beta particle dose absorbed in the immediate volume. The exceptions to this were in mutagenic studies with bacteria which had incorporated ^{3}H-uridine and ^{3}H-uracil. The incorporation into cytosine of these compounds revealed that decays at the 5-position were much more effective in producing damage than decays at 6-position. However, on the basis of the premise that only 2% of the DNA hydrogen is located at the 5-position of the cytosine ring, Carsten (1979) considered the risk was sufficiently small to be of no practical hazard in relation to the predicted effects of the emitted beta radiation. This conclusion was endorsed by Feinendegen and Bond (1971). Experimental studies of RBE would take account of such effects.

(ii) Contrary to the traditional view of 'organically bound tritium' in biomatter, recent experiments that employed denaturing agents have clearly shown that a significant form of such tritium should be designated as 'buried tritium', an exceedingly tightly bound tritiated water (Baumgartner and Donhaerl, 2004). In such a fraction the 'buried tritium' in biomacromolecules, such as native proteins, is in bridge positions where the exchange rates are reduced from microseconds to days, months or even years as a consequence of the three-dimensional structure that arises upon the 'folding' of these native biomacromolecules. The hydrogen bridges between the molecules of water are stronger than between organic configurations, resulting in accumulation of tritons both inside the biopolymers and within their primary hydration shields. There is an enrichment of tritium in the newly identified buried hydrogen bonds compared to the free water in the cell. In most biomacromolecules, e.g. proteins, the enrichment may be 1.4 x but in DNA, where the hydration shell consists of 11 molecules per nucleotide and is not readily permeable to ions, the enrichment in the water trapped in the core may be 2 x. While this will certainly result in slightly more beta tracks originating from tritiated water within and around the DNA, it remains true that the vast majority of beta tracks encountered by the DNA will have originated from tritiated water outside the DNA since that is where most of the tritiated water is situated. The effect on radiation dose to the DNA will therefore be small and, as stated above, should be seen as one of the factors tending to increase the RBE in experimental determinations. This has been emphasised in a recent paper (Melintescu et al, 2007).

4.2.1 Problems associated with RBE studies

4.2.1.1 Reference radiation

In the literature a range of reference radiations have been used for determination of RBE values for tritium and it is important when stating RBE values that the reference radiation is also specified. This is due to significant differences between LET values for typical reference radiations, such as ^{60}Co gamma rays and 200 kVp X-rays . Many experimental data over the years have shown that not all low LET radiations have the same effectiveness, especially at low doses. Orthovoltage X-rays are typically found to be twice as effective as high energy gamma rays at low doses, this difference is consistent with biophysical calculations (Ellett and Braby, 1972; Kellerer, 2002). Additionally differences have been observed for X-ray sets operated at the same potential but with different filtration. Similarly, the experimental values obtained for the RBE of tritium indicate differences in the biological effectiveness with reference to X-rays and gamma rays. In general, the values obtained when the reference radiation was ^{60}Co gamma rays were greater than the values obtained when the reference radiation was orthovoltage X-rays. There is currently no internationally agreed standard for the reference radiation but for a number of reasons (given below) a high energy gamma ray source would be the preferred choice for the reference radiation (ICRP, 2003) viz:

(a) Data for the atomic-bomb survivors represent the main source of information on radiation risks and these individuals were mainly exposed to high energy gamma rays.
(b) Most experimental animal studies of cancer induction have been carried out with gamma rays, including those at low dose rates.
(c) High energy gamma rays have the lowest values of LET.
(d) There is a more uniform distribution of energy deposition for large fields compared to X-rays.
(e) The LET and therefore the biological effectiveness of X-irradiations can depend on the filtration used.

4.2.1.2 Dose and dose rate

The RBE determined for tritium in any particular study will depend upon the shape of the dose–response curve for the endpoint of concern and the extent of the data available for both tritium and the reference low LET radiation to which the comparison is made.

In studies aimed at deriving a value of RBE for tritium there is the potential for the value obtained to depend upon the total dose and rate at which dose from both tritium and the reference radiation are delivered. In the case of tritium, the dose is expected to be protracted in time since the dose rate is dictated by the rate of radiological decay and rate of loss from the body. Experiments using X-rays or gamma rays, however, often deliver dose in a single acute exposure because this is more convenient. It is generally accepted that the same dose delivered in a protracted manner can have a lower effect than would an acute dose, due to the greater opportunity for DNA repair in the protracted case (ICRP, 1991; NCRP, 1980; NRC, 2006; UNSCEAR, 1993, 2000).

Most of the lower values for the RBE of tritium reported in the literature are from studies which have used higher doses and dose rates, particularly from the reference radiation. This trend can be explained by the high dose rate of the reference radiation reducing the apparent relative effectiveness of the tritium doses. A review of some tritium RBE studies was carried out by Ujeno (1983) which illustrates this phenomenon. Those studies which included external reference radiation showed a tendency for an inverse relationship between dose rate and RBE. The author concluded that use of a RBE of one would be reasonable for assessing the dose from very large intakes of tritium but that a figure larger than one would be more appropriate for environmental exposure situations. Therefore, the differences in the RBE with photon/electron energy can be viewed as differences in the DDREF assuming that the response at high acute doses is less dependent upon or is independent of photon/electron energy.

4.2.1.3 In vitro/in vivo studies

It is thought that the in vivo data are more appropriate because for practical radiation protection purposes the average organ dose will have been calculated following intake of tritium compounds and compared with that from an external radiation source. In general, RBEs for in vivo data would be expected on theoretical grounds to be less than for in vitro data. Track structures become more similar as radiation penetrates deeper into tissue, so that as more dose is due to 'deeper' tracks more of the dose is due to that from track ends (energy deposition at ends being less LET dependent). The experiments with chronically delivered comparison radiation are also more appropriate, given that tritium delivers dose in this way.

4.2.1.4 Concurrent reference radiation studies

It is generally considered important that, for RBE studies, the external reference radiation studies should have been carried out, if not at the same time, at least by the same lab. This is

because of genetic changes in animal strains and differences in methodology. Thus, in the data presented here we have excluded some studies considered by Straume and Carsten (1993) in their review that did not have adequate concurrent controls – for example, the studies of Carsten and Commerford (1976), Russell et al (1979), Prosser et al (1983) and Furuno-Fukushi et al (1987). Some of the calculations of RBE conducted by Straume and Carsten (1993) [e.g. the tritium experiments of Prosser et al (1983) combined with the gamma experiments of Lloyd et al (1975)] are based on non-concurrent experiments, and so should not be considered reliable.

4.2.1.5 HTO/OBT

Although experiments for determining the RBE of organically bound tritium (OBT) for practical radiation protection purposes, there are very few reliable studies – for instance, Carr and Nolan (1979), Lambert (1969) – and these concern tritiated thymidine which is of less importance in practical radiation protection.

4.2.2 Results from RBE studies

4.2.2.1 Straume and Carsten review

Straume and Carsten (1993) summarised the information then available on the radiobiological knowledge of carcinogenic, genetic, developmental and reproductive effects associated with exposures to tritium. They used these data to develop values for the RBE of tritium from about 40 papers which have been extensively quoted. Their overall assessment was that "tritium in the oxide form is about 2 to 3 times more effective at low doses or dose rates than gamma rays from ^{137}Cs or ^{60}Co."

4.2.2.2 Review of Kocher et al.

Some of these RBE data have been reviewed in a report by Kocher *et al* (2005). They described 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 to 5.0. It was not clear how they estimated the parameters of this distribution. They stated that "the upper tail of the distribution represents an assumption that [tritium RBE] could be 5 or greater, and that a reasonable upper bound cannot be determined with certainty". Nevertheless it should be noted that the distribution of RBE for tritium chosen by Kocher *et al*. is specifically for application to the calculation of probability of causation of cancer in compensation claims by radiation workers, and is thus likely to be pessimistic.

4.2.2.3 Carcinogenesis Studies

Potentially, animal studies which involved chronic or acute intakes of tritium with carcinogenesis as the endpoint would be most useful for the calculation of RBE for radiation protection purposes. Four such studies have been carried out (Gragtmans et al 1984, Johnson et al 1995, Seyama et al 1991, and Revina et al 1984) but the results should be treated with some care. For instance, a feature of the studies of Gragtmans , Revina and Seyama is the high background incidence of malignancy, so that in many cases what is measured is acceleration of onset rather than excess incidence.
These experimental animal carcinogenesis studies imply fairly modest tritium RBEs, with central estimates generally in the range 1.2 – 2.5, and an upper 97.5 percentile value of no more than about 3.0. Taking the three studies with adequate data (i.e., allowing an estimate of standard deviation) to compute the weighted RBE yields an aggregate RBE estimate with respect to chronic X-rays of 1.19 (95% CI 0.88, 1.49) [Gragtmans, and Johnson], and with respect to chronic γ rays of 2.49 (95% CI 2.00, 2.98) [Revina]. However, the shortcomings in

the experimental design (in one of them there is only a single dose point) and statistical analysis of these studies implies that, despite their obvious relevance to cancer, their findings should be treated with a degree of caution. Interpretation of all four studies is complicated by the fact that cancer incidence is near its maximum at the lowest doses.

4.2.2.4 Contemporary Reviews of the RBE of Tritium

Two recent reviews of tritium RBE studies have been made (AGIR 2007, Little and Lambert, 2007). These have further reviewed the published papers and updated them. Some of these data were deemed less reliable because they were not peer reviewed or were lacking concurrent reference radiation controls and have been excluded. This has not actually had a large effect on result of pooling the data. The general conclusions are:
"The distribution of RBEs among nine reliable studies in which X-rays were used as the comparison, the RBE distribution was highly skewed with most values in the range from one to two. In contrast, RBEs among the 14 studies for which gamma rays were used as the comparison were approximately normally distributed, with most values in the range from two to three. A more rigorous critical analysis of a restricted number of studies considered closest to being optimal has yielded an aggregate RBE estimate of 2.19 (95% CI 2.04, 2.33) with respect to chronic gamma radiation and 1.17 (95% CI 0.96, 1.39) with respect to chronic X-irradiation (Little and Lambert, 2007).
The RBEs summarised were obtained using radiations delivered at higher doses and dose rates than those generally received by workers or the public. Thus the average RBE values presented and the RBE distributions could, if anything, somewhat underestimate the actual RBEs most relevant for human risk assessment."

4.2.2.5 Overall conclusions with respect to Tritium RBE

(a) A variety of theoretical and experimental studies with radiation of LET similar to that of tritium beta particles has led to the general expectation of an RBE of at least two for tritium compared with gamma radiation.
(b) Neither transmutation effects nor isotopic discrimination associated with tritium appear likely to have a major effect, but any effect they might have would be in the direction of tending to increase the observed RBE.
(c) Experimentally determined RBE values can vary significantly depending on the choice of reference radiation but high energy gamma rays, should be the preferred choice for reporting RBE values. Where lower energy X-rays and gamma rays have been used as the standard, an appropriate adjustment should be taken into account when discussing the results. In addition, the reference radiation used should be adequately described including a description of anything that may modify the energy spectrum, such as filtration.
(d) Interpretation of RBE experiments is complicated by the fact that dose rates are rarely comparable and the reference radiation may itself be more effective than hard gamma rays.
(e) In a wide variety of cellular and genetic studies, RBEs for tritiated water have generally been observed in the range from one to two when compared with orthovoltage X-rays and in the range from two to three when compared with gamma rays.
(f) For developmental endpoints the RBEs for HTO were similar to those obtained for cellular and genetic studies.
(g) Whole animal carcinogenesis studies have yielded RBEs with central estimates generally in the range 1–2.5. However, there are several reservations about these studies. In particular, in some of the studies the frequency of cancers appears to have been saturated or nearly saturated at the lowest doses employed; the crucial low dose parts of the dose–response curves therefore cannot be compared.
(h) There are grounds for thinking that published RBEs could somewhat underestimate the actual RBEs relevant for human risk assessment since many of the studies employed

radiations delivered at higher doses and dose rates than those generally received by people.

(i) The most likely RBE relative to chronically delivered hard gamma rays is between two and three and a value of two is most appropriate, based largely on an analysis of the available experimental data with rounding and biophysical considerations; fractional values are probably not appropriate.

4.3 Epidemiology of Tritium

Clearly the most appropriate source of information for deriving risks to health from exposures to tritium would be from epidemiological studies if sufficient high quality data were available. However, the usefulness of such information for tritium is impaired by an almost total lack of dosimetric data, low doses and small numbers of cases. There have been several studies of both workers and members of the public who have, or might have been, exposed to tritium and these are reviewed below.

4.3.1 United Kingdom Atomic Energy Authority (UKAEA) workers

The study of Beral et al (1985) involved mortality follow-up of 39,546 UKAEA workers, from 1946 to 1979. The study of Atkinson et al (2004) involved mortality follow-up of a somewhat larger cohort, of 51,367 UKAEA workers , extending follow-up to the end of 1997. There was a statistically significant elevation in prostate cancer mortality in comparison with national rates in the cohort of 1416 workers monitored for tritium in the study of Beral et al (1985): the standardised mortality ratio (SMR) was 8.89 (2-sided $p<0.001$), based on 6 deaths; there were no statistically significantly elevated SMRs for nine other endpoints. There are no data given on tritium doses to this group (nor for doses from any other internally deposited radionuclide). The study of Rooney et al (1993) involved follow-up of UKAEA workers for prostate cancer before 1986.

Data for a total of 136 cases of prostate cancer and 404 controls (comprised of 372 individuals, some matched to more than one case) were collected. The study assessed possible exposure of each case or control to a number of radionuclides, including tritium, on a four-point scale (none, possible, probable

but low level, probable, and relatively high level). Rooney et al (1993) found a relative risk of 14.26 (95% CI 3.09, 133.16) associated with documented internal contamination by tritium, and an increasing trend (2-sided $p<0.05$) of prostate cancer risk with degree of potential exposure to tritium. The main

problem with this study is a lack of adequate tritium dosimetry, so that interpretation of these figures to derive risk estimates for tritium is problematic.

4.3.2 Atomic Weapons Establishment (AWE) workers

The study of Beral et al (1988) involved mortality follow-up of 22,552 AWE workers from 1951 to 1982. There are no data given on tritium doses to this group (nor for doses from any other internally deposited radionuclide). Among 1562 workers ever monitored for tritium, and among 20 causes of death considered there was a statistically significant (2-sided $p<0.01$) elevation in brain and central nervous system mortality relative to other radiation workers, based on a single case; there is no statistically significant trend with dose in the full cohort. In the absence of tritium-specific doses it is difficult to infer very much about risk from tritium in this study; the apparently significant findings in relation to prostate and brain cancer should be considered against the negative findings for these endpoints in the study of Carpenter et al (1998), which analysed this workforce and various other UK radiation workforces.

4.3.3 Three groups of UK classified workers

The study of Carpenter et al (1998) involved follow-up of three groups of UK classified radiation workers, employed at the UKAEA establishments at Harwell (including Culham and London), Dounreay or Winfrith before 1980, the AWE before 1983, and at Sellafield before 1976. A total of 40,751 workers were studied, of whom 4111 were monitored for exposure to tritium. There are no data given on tritium doses to this group (nor for doses from any other internally deposited radionuclide); all analyses are in terms of cumulative external dose. There is little evidence of a raised risk associated with tritium exposure, but in the absence of tritium-specific doses it is difficult to infer very much about risk from tritium.

4.3.4 Savannah River workers and other US nuclear workers

Cragle et al (1988) assessed mortality in a cohort of 9860 white male workers at the Savannah River Site, South Carolina, USA, from 1952 to 1980. A further analysis of this cohort by Cragle et al (1998) extended follow-up to 1986, but has not been published in the peer-reviewed literature. There are few data given on tritium doses to this group (nor for external doses nor from any other internally deposited radionuclide), although there are indications that the workforce was relatively heavily exposed to tritium. About 800 employees received 0.5 mSv from tritium, and one worker received a dose of over 30 mSv from this source per year (Cragle et al, 1988). There are few indications of excess mortality in this cohort; in particular, there are 8 prostate cancer deaths versus 10.84 expected (SMR=0.74), although there are stronger (but still not statistically significant) indications of an excess for leukaemia and aleukaemia (18 deaths versus 13.29 expected, SMR=1.35) (Cragle et al, 1988). In the absence of tritium-specific doses it is difficult to infer very much about risk from tritium in this study.
Schubuer-Berrigan et al (2007) assessed acute leukaemia and chronic myeloid leukaemia mortality at four nuclear sites in the USA, i.e. Hanford, Oak Ridge National Laboratories, Savannah River Site and Los Alamos National Laboratories, together with the Portsmouth Naval Shipyard. A nested case–control design within the respective cohorts was employed. Various periods of follow-up were used for the various component cohorts, generally finishing between 1994 and 1996. A total of 206 leukaemia cases and 823 age-matched controls were used. Dose estimation included that from tritium, as well as photons, neutrons and plutonium. The unadjusted excess odds ratio (EOR) for the Savannah River Site workers, likely to be among the most heavily exposed to tritium among these five workforces, was 30.6 Sv–1 (95% CI 4.77, >130). There was no analysis accounting separately for the effects of tritium and other radiation exposures, and in the absence of this it is difficult to infer much about tritium risks from these three studies. However, the Savannah River Site workers are potentially informative about tritium risks.

4.3.5 Capenhurst uranium enrichment workers

At the Capenhurst uranium enrichment facility tritium gas was processed between 1965 and 1987 for defence purposes (Jackson et al, 1997). It is not clear how many of the 12,540 Capenhurst workers assessed in the report of McGeoghegan and Binks (2000) were exposed to tritium; tritium doses were not available. There were no statistically significant positive trends of cancer mortality with cumulative external dose. The only statistically significant positive trend (1-sided $p<0.05$) of cancer morbidity with cumulative external dose was for bladder cancer (out of 16 cancer sites examined), based on 14 cases. The trend of prostate cancer with cumulative external dose was negative. In the absence of tritium-specific doses it is difficult to infer very much about risk from tritium in this study.

4.3.6 Sellafield workers

There are tritium-exposed workers at the Sellafield site, although they have never been separately assessed, nor are there analyses in relation to tritium dose, for example in the analyses by Omar et al (1999) or McGeoghegan et al (2003). There are no statistically

significant trends with cumulative external radiation dose in the analysis of the female workers (McGeoghegan et al, 2003).
Tritium-specific doses have been estimated for this workforce, although they have not been taken into account in the analyses discussed here. In the absence of analysis in which these are explicitly taken into account it is difficult to infer very much about the risk from tritium in these studies, although this cohort should be regarded as potentially informative.

4.3.7 Canadian nuclear workers

The study of Zablotska et al (2004) involved mortality follow-up of 45,468 Canadian nuclear workers between 1957 and 1994. Tritium doses were calculated from urinalysis data for all workers who had potential exposure, and added to external (film-badge) doses. There is no indication in the published analyses of the magnitude of the tritium doses, but for some workers they are likely to be relatively substantial. Overall, this cohort had a mean dose of 13.5 mSv, and among those workers recorded as having some dose the mean was 19.7 mSv. As with many studies of nuclear workers the expected numbers were below national rates. For example, this was the case for all cancers (531 observed versus 721.0 expected, SMR=0.72, 95% CI 0.68, 0.80) and leukaemia excluding CLL (18 observed versus 22.6 expected, SMR=0.80, 95% CI 0.47, 1.26).

4.3.8 Conclusions from studies of radiation workers

(a) Tritium-specific doses have been estimated for some nuclear industry workforces, in particular those of Sellafield and the AWE in the UK, and in Canada.
(b) Only for the Canadian (Zablotska et al, 2004), American (Schubauer-Berrigan et al, 2007) and AWE workers (Johnson et al, 1999) are these doses explicitly taken into account in the analysis, although not in a way that facilitates inferences on risks associated with tritium.
(c) All of the workforces considered are likely to have some workers relatively highly exposed to tritium.
(d) Some nuclear industry workforces, in particular those at the Savannah River Site (Cragle et al 1988, 1998; Schubauer-Berrigan et al, 2007), and the Canadian workers (Zablotska et al, 2004), have relatively large numbers of workers with appreciable tritium exposures (relative to the other groups), and would be potentially informative.

4.3.9 Offspring of Canadian electric power workers

Green et al (1997) assessed cases of congenital abnormalities and matched controls in the offspring of Canadian electric power workers, specifically offspring of Ontario Hydro workers. There were 763 case–control pairs of fathers and 165 case–control pairs of mothers. Tritium doses were assessed for this group, although analyses were only of those cases and controls with fathers or mothers having a recorded tritium dose 60 days before conception versus those with no dose. There is little evidence of a raised risk associated with tritium exposure.

4.3.10 Offspring of Ontario radiation workers

McLaughlin et al (1992, 1993) considered cases of childhood (age 0–14 years) leukaemia in the offspring of Ontario radiation workers and matched cases. Workers were those employed at the AECL laboratories at Chalk River, a uranium processing plant at Port Hope, a uranium mining and milling plant at Elliot Lake and five power reactors (Rolphton, Pickering A and B, and Bruce A and B). There were 112 cases and 896 controls. Preconceptional tritium doses were assessed for this group. Again, there is little evidence of a raised risk associated with tritium exposure.

4.3.11 Leukaemia in children in the vicinity of Kruemmel and Savannah River

Grosche et al (1999) studied cases of childhood leukaemia in the vicinity of the Kruemmel nuclear power plant in Germany, and the Savannah River Site in the USA. They observed 9 cases of childhood leukaemia in the period 1990–1996 within 10 km of the Kruemmel plant (standardised incidence ratio, SIR=3.25, 95% CI 1.58, 5.96); over the period 1991–1995 Grosche et al observed 41 cases of childhood leukaemia in the Savannah River Region Health Information System (SIR 0.86, 95% CI 0.59, 1.21). Although there is no individual dosimetry for either group, Grosche et al pointed out that tritium releases from the Savannah River plant exceed by several orders of magnitude those from the Kruemmel plant, so that the apparent excess leukaemia risk near the Kruemmel plant is probably not associated with tritium exposure.

4.3.12 Leukaemia in children in the vicinity of Canadian nuclear facilities

Clarke et al (1989, 1991) examined mortality and incidence of childhood leukaemia in the vicinity of nuclear facilities in Ontario. The first report (Clarke et al, 1989) considered leukaemia deaths and cases at ages 0–4 years, and the second report (Clarke et al 1991) considered cases and deaths at ages 0–14 years. Overall there is no evidence of excess leukaemia incidence or mortality near the six power reactor sites or the other nuclear installations

4.3.13 Birth defects and infant mortality in the vicinity of the Pickering nuclear facility, Ontario

Johnson and Rouleau (1991) studied birth defects, stillbirths, and perinatal, neonatal and infant mortality within 25 km of the Pickering nuclear generating facility in Ontario. There is no overall evidence of excess mortality near Pickering – if anything rates were significantly less than the Ontario average. Johnson and Rouleau (1991) also studied these endpoints in relation to airborne and waterborne discharges of tritium from the Pickering plant, concentrating on the Pickering and Ajax townships, those closest to the plant. The incidence of central nervous system defects was significantly elevated in Pickering for airborne discharges at the highest of five levels (odds ratio, OR, in highest group = 4.01, 95% CI 1.25, 14.04, based on 6 cases), but there was no statistically significant trend with tritium exposure (p=0.197). However, when the analysis was repeated using Health and Welfare ground monitoring data for tritium there was no association for this endpoint (or any other) (OR for CNS defects in the highest exposure group = 0.24). There was a statistically significantly raised prevalence of births with Down's syndrome in Pickering (24 observed versus 12.9 expected (relative risk, RR=1.85, 95% CI 1.19, 2.76), the only endpoint out of 23 birth defect endpoints to show such an excess. There was a correlation, although not statistically significant (p=0.468), between Down's syndrome prevalence and airborne tritium release, but no such correlation with Health and Welfare ground monitoring data. There were no such excess birth defect risks in Ajax, although there was a positive correlation, but again not statistically significant (p=0.282), with Health and Welfare ground monitoring data. As the authors pointed out, the few positive findings in this study are likely to be due to chance. The ecological design of the study is also a potential source of bias, firstly because airborne release levels in an area are poor indicators of individual exposure and secondly because geographical confounding factors may influence the relationship between outcomes such as congenital abnormalities and area-averaged tritium releases. A similar study of other nuclear power stations in Ontario does not appear to have been carried out.

4.3.14 Overall conclusions from epidemiological studies

(a) In general, the available epidemiological studies do not contain enough detail to estimate risks from tritium.

(b) Of the populations identified, the best prospects for more robust analyses exist in the nuclear worker studies in Canada (Zablotska et al, 2004), Savannah River Site (Cragle et al, 1988, 1998) and possibly other sites in the USA, and the five main tritium-exposed UK nuclear workforces, namely those at Sellafield, Chapelcross and Capenhurst and at the AWE and the UKAEA.
(c) All of these workforces have some groups who are relatively highly exposed to tritium, and with apparently good dosimetry. These could be used as the basis of a further study and this should be encouraged.

4.4 References

AGIR (2007) Report of Independent Advisory Group on Ionising Radiation. Review of risks from tritium. Docs of HPA. RCE-4 .

Atkinson WD, Law DV, Bromley KJ and Inskip HM (2004). Mortality of employees of the United Kingdom Atomic Energy Authority, 1946–97. Occup Environ Med, **61**, 577–85.

Baumgartner F and Donhaerl W (2004). Non-exchangeable organically bound tritium (OBT): its real nature. Anal Bioanal Chem, **379**, 204–9.

Beral V, Inskip H, Fraser P, Booth M, Coleman D and Rose G (1985). Mortality of employees of the United Kingdom Atomic Energy Authority, 1946–1979. Br Med J, **291**, 440–47.

Bouffler SD, Bridges BA, Cooper DN, Dubrova Y, McMillan TJ, Thacker J, Wright EG and Waters R (2006). Assessing radiation-associated mutational risk to the germline: repetitive DNA sequences as mutational targets and biomarkers. Radiat Res, **165**(3), 249–68.

Bridges BA (2001). Radiation and germline mutation at repeat sequences: are we in the middle of a paradigm shift? Radiat Res, **156** (5 Part 2), 631–41.

Carpenter LM, Higgins D, Douglas AJ, Maconochie NES, Omar RZ, Fraser P, Beral V and Smith PG (1998). Cancer mortality in relation to monitoring for radionuclide exposure in three UK nuclear industry workforces. Br J Cancer, **78**, 1224–32.

Carr TEF and Nolan J (1979). Testis mass loss in the mouse induced by tritiated thymidine, tritiated water and 60Co gamma irradiation. Health Phys, **36**, 135–45.

Carsten AL (1979). Tritium in the environment: isotopic effects and transmutation. In Advances in Radiation Biology, **8,** 419–58 (JT Lett and H Adler, eds). New York, Academic Press.

Carsten AL and Commerford SL (1976). Dominant lethal mutations in mice resulting from chronic tritiated water (HTO) ingestion. Radiat Res, **66,** 609–14.

Clarke EA, McLaughlin J and Anderson TW (1989). Childhood Leukaemia around Canadian Nuclear Facilities – Phase I. Final report. Report INFO-0300. Ottawa, Atomic Energy Control Board.

Clarke EA, McLaughlin J and Anderson TW (1991). Childhood Leukaemia around Canadian Nuclear Facilities –Phase II. Final report. Report INFO-0300-2. Ottawa, Atomic Energy Control Board.

Cleaver JE (1977). Induction of thioguanine- and ouabain-resistant mutants and single-strand breaks in the DNA of Chinese hamster ovary cells by 3H-thymidine. Genetics, **87,** 129–38.

Cragle DL, McLain RW, Qualters JR, Hickey JLS, Wilkinson GS, Tankersley WG and Lushbaugh CC (1988). Mortality among workers at a nuclear fuels production facility. Am J Indust Med, **14**, 379–401.

Cragle DL, Watkins JP and Robertson-DeMers K (1998). Mortality among workers at the Savannah River nuclear fuels production facility. In Proceedings of the Section on Statistics in

Epidemiology. Alexandria VA, American Statistical Association, pp83–87. URL: www.orau.gov/ehsd/EpiProceedings-jj-9-30-98.doc.

DeLara CM, Hill MA, Jenner TJ, Papworth D and O'Neill P (2001). Dependence of the yields on DNA double-strand breaks in Chinese hamster V79-4 cells on the photon energy of ultrasoft X-rays. Radiat Res, **155**, 440–48.

EA, EHSNI, FSA and SEPA (2006). Radioactivity in Food and the Environment, 2005. RIFE 11. Lowestoft, Centre for Environment, Fisheries and Aquaculture Science.

Ellett WH and Braby LA (1972). The microdosimetry of 250 kVp and 65 kVp X-rays, 60Co gamma rays, and tritium beta particles. Radiat Res, **51**, 229–43.

Feinendegen LE (1967). Problems associated with the use of labelled molecules in biology and medicine. In Proceedings of Biological Effects of Transmutation and Decay of Incorporated Radioisotopes, pp1–15, Vienna, IAEA.

Feinendegen LE and Bond VP (1971). Transmutation versus beta irradiation in the pathological effects of tritium decay. In Tritium (AA Moghissi and MW Carter, eds). Nevada, Messenger Graphics, pp221–31.

Forell B, Myers LSJ and Norman A (1982). DNA repair synthesis in minimally stressed human lymphocytes. Intern J Radiat Biol, **41**, 535–45.

Furono-Fukushi I, Ueno AM and Matsudaira H (1987). Cell killing and mutation to 6-thioguanine resistance after exposure to tritiated amino acids and tritiated thymidine in cultured mammalian cells (L5178Y). Radiat Res, **110**, 428–38.

Gragtmans NJ, Myers DK, Johnson JR, Jones AR and Johnson LD (1984). Occurrence of mammary tumors in rats after exposure to tritium beta rays and 200-kVp X-rays. Radiat Res, **99**, 636–50.

Green LM, Dodds L, Miller AB, Tomkins DJ, Li J and Escobar M (1997). Risk of congenital anomalies in children of parents occupationally exposed to low level ionising radiation. Occup Environ Med, **54**, 629–35. 62

Grosche B, Lackland D, Mohr L, Dunbar J, Nicholas J, Burkart W and Hoel D (1999). Leukaemia in the vicinity of two tritium-releasing nuclear facilities: a comparison of the Kruemmel Site, Germany, and the Savannah River Site, South Carolina, USA. J Radiol Prot, **19**, 243–52.

Hodgson A, Scott JE, Fell TP and Harrison JD (2005). Radiation doses from the consumption of Cardiff Bay flounder containing organically bound tritium (OBT). J Radiol Prot, **25,** 149–59.

ICRP (1991). Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Ann ICRP.

ICRP (2003). Relative biological effectiveness (RBE), quality factor (Q), and radiation weighting factor (w R). ICRP Publication 92. Ann ICRP.

Jackson D, Gray J, Powell F and Roscoe PA (1997). Reconstruction of discharges and environmental doses from a former tritium facility at BNFL Capenhurst. J Radiol Prot, **17**, 239–52.

Johnson JR, Myers DK, Jackson JS, Dunford DW, Gragtmans NJ, Wyatt HM, Jones AR and Percy DH (1995). Relative biological effectiveness of tritium for induction of myeloid leukaemia in CBA/H mice. Radiat Res, **144**, 82–9.

Johnson KC and Rouleau J (1991). Tritium Releases from the Pickering Nuclear Generating Station and Birth Ddefects and Infant Mortality in Nearby Communities 1971–1988 (AECB Project No. 7.156.1). Report INFO-0401. Ottawa, Atomic Energy Control Board.

Johnson P, Atkinson WD and Nicholls JL (1999). Updated analysis of mortality in workers at UK atomic weapons establishments. In Achievements and Challenges: Advancing Radiation

Protection into the 21st Century. Proceeding 6th SRP International Symposium, Southport, June 1999 (MC Thorne, ed). London, Society for Radiological Protection.

Kacena V (1967). Chemical effects of decay of incorporated radioisotopes. In Proceedings Conference on Biological Effects of Transmutation and Decay of Incorporated Radioisotopes. Vienna, IAEA, pp199–214.

Kellerer AM (2002). Electron spectra and the RBE of X-rays. Radiat Res, **158**, 13–22.

Kocher DC, Apostoaei AI and Hoffman FO (2005). Radiation effectiveness factors for use in calculating probability of causation of radiogenic cancers. Health Phys, **89**, 3–32.

Korolev VG and Ivanov FL (1985). Mutagenic effect and mutation spectrum induced by 3H decay in the 8th position of DNA purine bases in Saccharomyces cerevisiae. Mutat Res, **149,** 359–64.

Krasin F, Person S, Ley RD and Hutchinson F (1976). DNA crosslinks, single-strand breaks and effects on bacteriophage T4 survival from tritium decay of (2-3H)adenine, (8-3H)adenine and (8-3H)guanine. J Mol Biol, **101,** 197–209.

Krisch RE and Zelle MR (1969). Biological effects of radioisotope decay: the role of the transmutation effect. In Advances in Radiation Biology (LG Augenstein et al, eds). New York, Academic Press, **3,** pp177–213.

Lambert BE (1969). Cytological damage produced in the mouse testes by tritiated thymidine, tritiated water and X-rays. Health Phys, **17**, 547– 57.

Little MP and Lambert BE (2007). Systematic review of experimental studies on the relative biological effectiveness of tritium. Radiat. Environ. Biophys. [in press]

LLNL (2002). Tritium Releases and Potential Offsite Exposures. The Lawrence Livermore National Laboratory and the Savannah River Site: Agency for Toxic Substances and Disease Registry (ATSDR) Health Consultation Report.

Lloyd DC, Purott RJ, Dolphin GW, Bolton D, Edwards AA and Corp MJ (1975). The relationship between chromosome aberrations and low LET radiation dose to human lymphocytes. Int J Radiat Biol, **28**, 75–90.

McGeoghegan D and Binks K (2000). The mortality and cancer morbidity experience of workers at the Capenhurst uranium enrichment facility, 1946–95. J Radiol Prot, **20**, 381–401.

McGeoghegan D, Gillies M, Riddell AE and Binks K (2003). Mortality and cancer morbidity experience of female workers at the British Nuclear Fuels Sellafield plant, 1946–1998. Am J Indust Med, **44**, 653–63.

McLaughlin J, Anderson TW, Clarke EA and King W (1992). Occupational Exposure of Fathers to Ionizing Radiation and the Risk of Leukaemia in Offspring – A Case–control Study (AECB Project No. 7.157.1). Report

INFO-0424. Ottawa, Atomic Energy Control Board.

McLaughlin JR, King WD, Anderson TW, Clarke EA and Ashmore JP (1993). Paternal radiation exposure and leukaemia in offspring: the Ontario case–control study. Br Med J, **307**, 959–66, Errata: **307**, 1257, 1462.

Masui Y and Pedersen RA (1975). Ultraviolet light-induced unscheduled DNA synthesis in mouse oocytes during meiotic maturation. Nature (London), **257**, 705–6.

Melintescu A, Gateriu D and Takeda H (2007). Reassessment of tritium dose coefficients for the general public. Rad. Prot. Dosim. 1-5

NCRP (1979). Tritium and Other Radionuclide Labelled Organic compounds incorporated in Genetic Material. NCRP Report No 63. Washington DC, National Council on Radiation Protection and Measurements.

NCRP (1980). Evaluation of the Linear Nonthreshold Dose–response Model for Ionizing Radiation. NCRP Report No. 64. Washington DC, National Council on Radiation Protection and Measurements.

NCRP (2001). Influence of Dose and its Distribution in Time on Dose–response Relationships for Low-LET Radiation. NCRP Report No. 136. Bethesda MD, National Council on Radiation Protection and Measurements.

NRC (2006). Committee to Assess Health Risks from Exposure to Low Levels of Ionizing Radiation. Health Risks from Exposure to Low Levels of Ionizing Radiation: BEIR VII – Phase 2. National Academy of Sciences/National Research Council. Washington DC, National Academy Press.

Omar RZ, Barber JA and Smith PG (1999). Cancer mortality and morbidity among plutonium workers at the Sellafield plant of British Nuclear Fuels. Br J Cancer, **79**, 1288–301.

Pedersen RA and Mangia F (1978). Ultraviolet-light-induced unscheduled DNA synthesis by resting and growing mouse oocytes. Mutat Res, **49**, 425–9.

Pelc SR (1968). Turnover of DNA and function. Nature (London), **219**, 162–3.

Person S, Snipes W and Krasin F (1976). Mutation production from tritium decay: a local effect for (3H)α-adenosine and (3H)6-thymidine decays. Mutat Res, **34**(2), 327.

Prosser JS, Lloyd DC, Edwards AA and Stather JW (1983). The induction of chromosome aberrations in human lymphocytes by exposure to tritiated water. Radiat Prot Dosim, **4,** 21–6.

Revina VS, Voronin VS, Lemberg VK, and Sukhodoev VV (1984). Comparative evaluation of the tumorigenic effect of chronic exposure to tritium oxide and external gamma radiation. Radiobiologiia, **24**, 697–700.

Rooney C, Beral V, Maconochie N, Fraser P and Davies G (1993). Case–control study of prostatic cancer in employees of the United Kingdom Atomic Energy Authority. Br Med J, **307**, 1391–7.

Russell WL, Cumming RB, Kelly EM and Phipps EL (1979). Induction of specific-locus mutations in the mouse by tritiated water. In Behaviour of Tritium in the Environment. Proceedings Symposium on the Behaviour of

Tritium in the Environment, October 1978, San Francisco CA, USA, pp489–97. Vienna, IAEA, IAEA-SM 232/85.

Seyama T, Yamamoto O, Kinomura A and Yokoro K (1991). Carcinogenic effects of tritiated water (HTO) in mice: In comparison to those of neutrons and gamma-rays. J Radiat Res, Supplement 2, 132–42.

Straume T and Carsten AL (1993). Tritium radiobiology and relative biological effectiveness. Health Phys, **65**, 657–72.

Teebor GW, Frenkel K and Goldstein MS (1984). Ionizing radiation and tritium transmutation both cause formation of 5-hydroxymethyl-2'-deoxyuridine in cellular DNA. Proc Natl Acad Sci USA, **81**(2), 318–21.

Tisljar-Lentulis G, Henneberg P, Feinendegen LE and Commerford SL (1983). The oxygen enhancement ratio for single- and double-strand breaks induced by tritium incorporated in DNA of cultured human T1 cells. Impact of the transmutation effect. Radiat Res, **94,** 41–50.

Ujeno Y (1983). Relative biological effectiveness (RBE) of tritium β rays in relation to dose rate. Health Phys, **45,** 789–91.

UNSCEAR (1993). The influence of dose and dose rate on stochastic effects of radiation. In Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation Report to the General Assembly, with scientific annexes. Vienna, United Nations, pp619–728.

UNSCEAR (2000). Sources and Effects of Ionizing Radiation. Volume I: Sources; Volume II: Effects. United Nations Scientific Committee on the Effects of Atomic Radiation Report to the General Assembly, with scientific annexes. New York, United Nations.

Williams JL, Russ RM, McCubbin D, and Knowles JF (2001). An overview of tritium behaviour in the Severn Estuary (UK). J Radiol Prot, **21,** 337–44.

Yamada T, Yukawa O, Asami K and Nakazawa T (1982). Effect of chronic HTO beta or ^{60}Co gamma radiation on preimplantation mouse development in vitro. Radiat Res, **92**, 359–69.

Zablotska LB, Ashmore JP and Howe GR (2004). Analysis of mortality among Canadian nuclear power industry workers after chronic low-dose exposure to ionizing radiation. Radiat Res, **161**, 633–41.

5 TRITIUM AND THE ENVIRONMENT: SOURCES, MEASUREMENT AND TRANSFER

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5.1 Abstract

This document presents the different sources of tritium, different methods of measurement associated to tritium processes and survey, transfer in the biosphere compartments in normal and accidental conditions. Some R&D subjects are suggested and some regulatory aspects are discussed.

5.2 Sources of tritium

Tritium (^{3}H) in the environment has two origins, natural and man-made.

Tritium is produced mainly as the result of interaction of cosmic neutrons on nitrogen, oxygen and argon. About 99% are incorporated in water and follow the water cycle from the high atmosphere to the sea. A global natural inventory at about $1.3.10^{18}$ Bq (1300 PBq or 3.5 kg) has been evaluated by UNSCEAR [UNSCEAR 2000].

The annual production of tritium ranges from 50 to 70 PBq (0.15 to 0.20 kg).

The ^{3}H from man-made origin comes from many sources:

- Far above all other sources the tritium is released during the **atmospheric tests** in the decades 50 and 60, in both hemispheres (respectively 420 kg in the north and 140 kg in the south). From 1945 to 1963, these tests released about 560 kg of tritium. Considering radioactive decay of tritium, it remained about 40 kg in 2007, distributed for nine tenths in the sea, one tenth in continental waters and about 1% in the atmosphere.
- The second source is the **spent fuel reprocessing plants**. Most of the processing plants are near the sea and releases are essentially liquid. La Hague releases about 10 $PBq.y^{-1}$ (30g) for 1600 t of material [Carmin 2005], and Sellafield about 2 or 3 PBq (8g).

These sources (atmospheric releases of respectively 0.07 and 0.2 PBq) have a little effect on atmosphere.

Nuclear reactors have different tritium production depending on the type of reactor.

In a **PWR** [Jones 2007], tritium is produced primarily from neutron capture by ^{10}B. Boric acid is added to PWR reactor coolant system (RCS) as a soluble reactivity shim, and boron (enriched in ^{10}B) is used in fuel assemblies as a burnable poison. 90% of the total tritium in PWR reactor coolant are produced in the coolant by the soluble boric acid reactivity shim. The remaining 10% are produced by ternary fission, ^{10}B burnable poisons, ^{6}Li neutron capture, and deuterium activation. Neutron capture by ^{7}Li is a minor tritium source.
The use of boron (chemical shim) increases: early core designs used other poisons; most PWR's have extended refuelling cycle from 18 to 24 months; longer cycles require higher initial boron concentration in reactor coolant; core power upgrades require more boron. Boron increase causes tritium increase.

Atmospheric releases of PWR are about 0.3 to 3 $TBq.y^{-1}$ in the USA. Liquid releases have increased from 10 to 20 $TBq.y^{-1}$ between 1973 and 2006. UNSCEAR [UNSCEAR 2000] gives the same ranges of values.

BWR tritium production is significantly lower than PWR due to absence of boric acid in the coolant. Airborne effluents releases are about 1 $TBq.y^{-1}$ and equally for liquids.

GCR -Gas Cooled Reactors- liquid releases are about 10 times higher.

HWR - Heavy Water Reactors – These reactors produce tritium from deuterium: it explains why there are much higher releases. Airborne effluents are about 100 to 1000 $TBq.y^{-1}$ and liquid releases also about 100 to 500 $TBq.y^{-1}$(about 1g).

5.2.1 Industrial and small users

Two types of industrial use have to be noticed: one is the production and use of labelled compounds for medical or R&D purposes. Amersham, in Great Britain, released 0.5 PBq.y-[1] (airborne effluent) before 2000 and reduced to 0.1 $PBq.y^{-1}$. Releases of dissolved organic compounds have "labelled" the food chain of the estuary environment with less dilution than expected [Wallis 2005]. The second is the production by gaseous tritium light facilities of tritiated devices in Canada which have released 1.6 $PBq.a^{-1}$ in 2000 with a decrease until 2006 down to 0.1 PBq of HTO (tritiated water) and 0.3 PBq of HT (tritium gas) [Mihok 2008]. This type of devices seems to be sources of measurable tritium in landfills, like in USA [Mutch 2008] where recent studies show some tritium in water table values higher than the 20000 pCi quality standard for most of the states. One EXIT lighting device may contain 0.1 to 0.5 TBq.

Small users of labelled compounds, hospitals, pharmaceutical laboratories produce few releases, and the stock of waste is evaluated in France at 0.5 PBq. Most of the present production of wastes can be incinerated or disposed in surface disposal.

5.2.2 Future use

Future use concern few $mg.a^{-1}$ in laser like Laser MegaJoule, and mainly fusion reactors. The Joint European Torus used 20g. ITER will use 1.5 $kg.y^{-1}$. Releases are presently not definitively defined. ITER safety report indicates 1850 $TBq.y^{-1}$ (5g) gas release.

5.2.3 Waste

Between 1967 and 1982, about 20 PBq of tritium wastes have been eliminated by sea dumping. In France, the radiological capacity of the Low-Medium-Level-Waste disposal centre of Aube is 4 PBq, but acceptance requirements for tritiated waste are very difficult to meet. In practice pure tritium waste are not sent to the Aube Centre. The Very-Low-Level-Waste disposal centre has also very low acceptance criteria.
Some waste, because they contain other nuclides than tritium, have to be managed in deep geological or subsurface disposal (type B). Nevertheless they can contain significant amount of tritium. This is the case of graphite in France: in 2007, 20000 t contained less than 5 PBq, which will probably be disposed in subsurface disposal. It is also the case of hules and nozzles that can contain $20TBq.t^{-1}$ and will be disposed in geological disposal. Presently outgassing of these waste appears to be very small, in the range of 10^{-6} to 10^{-7} y^{-1}. Same values may be noticed for B_4C rods of reactors.

In 2007, French military application waste storage represented about 5 PBq. The policy is to treat all the waste coming from the first barrier of process by fusion of metals and vapor treatment for organic waste with large outgassing rate. Outgassing rate is of about 1% per year [Batifol 2008]. Outgassing rate clearly depends on temperature, as shown in figure 1.

Figure 2 gives an overview of authorized airborne and liquid tritium releases in France and some real releases from a Candu Canadian reactor.

Figure 3 gives an evaluation of tritiated waste production in France in the next 50 years.

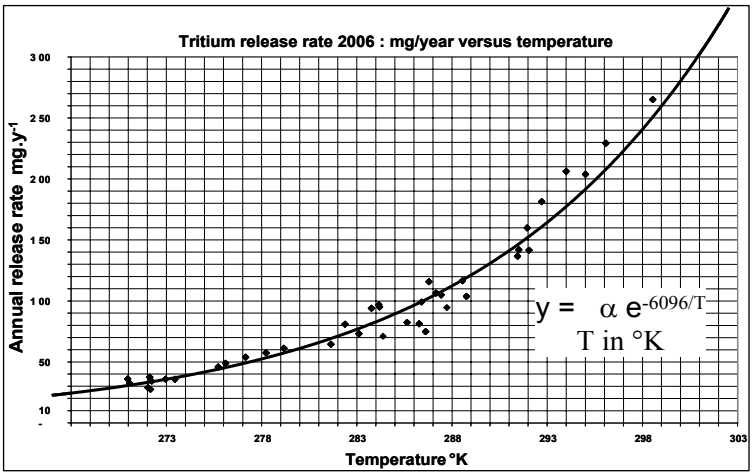

Figure 1: Tritium release rate (mg.y^{-1}) versus Temp. °K: in a Waste Storage

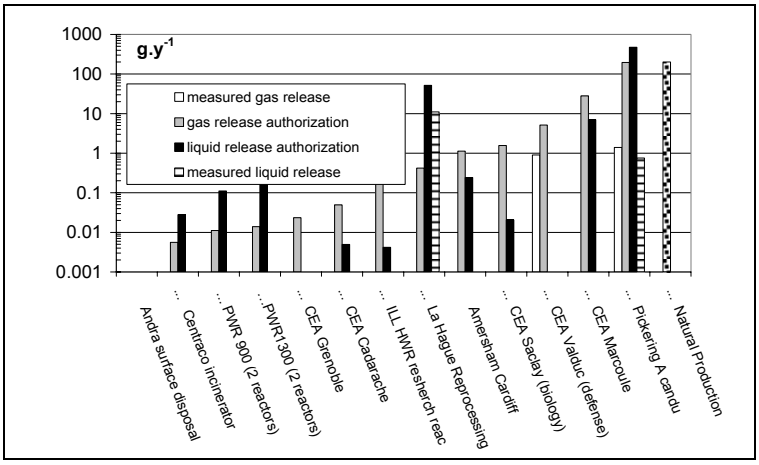

Figure 2: Different levels of authorized releases of tritium [gram]

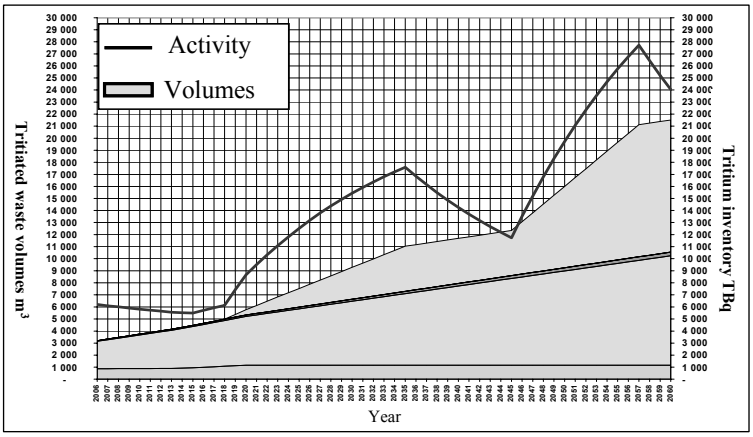

Figure 3: Prospective cumulated volumes and tritium activities of tritium waste in the next 50 years.

The increase justifies the requirement of the Program law N° 2006-739 28th June 2006 concerning durable management of radioactive waste and material: " … it is defined a R&D program with the following objective: … find up to the end of 2008 storage solutions for waste containing Tritium so that radioactivity decay allows their disposal in surface or shallow disposal."

5.3 Measurement

Tritium may be in very different forms: as a gas, it can be dihydrogen H_2, water vapour H_20, methane CH_4. It may be pure or with different isotopies (^1H, ^2H, ^3H) and contains impurities, at least ^3He.

As a liquid, it comes often as water, but can also be dissolved organic compounds, scintillation cocktail, solvents or oils…

Tritium can also be in the solid form: in metal where diffusion occurs; in any solid organic compounds, it can also be powder of uranium, palladium… or hydride which are or can be used for a solid storage of hydrogen. It can also come as water adsorbed on zeolithe.

It covers the range of activities from traces in surface water 0.1 $Bq.kg^{-1}$ to pure tritium 3.58 10^{17} $Bq.kg^{-1}$.

Before atmospheric tests, concentrations in rain, rivers and oceans were respectively 0.6 $Bq.L^{-1}$, 0.3-0.8 $Bq.L^{-1}$, < 0.1 $Bq.L^{-1}$. Rain concentration reached 150 $Bq.L^{-1}$ in 1963 in north hemisphere and has been decreasing since that time to about 1 $Bq.L^{-1}$ presently.

Surface water	0,1-0,9
Rain water (before tests)	0,6
Rain water (1963)	150
Rain water (1990)	2
Aquifer	1 - 10
Rivers	<10
North sea	0.3
Oceans : surface	0.1
Deep -1000m	0.005

Table 1: Tritium Concentrations in natural waters ($Bq.L^{-1}$).

It is important to keep in mind that air naturally contains tens of grams of water vapor, that tritium (gas or water) is very mobile and may be a source of contamination from samples to other samples.

Tritium measurements are used for many purposes: inventories, quality (isotopy and impurities) releases, characterization of waste (total activity and outgassing), and control of the environment for normal or accidental releases. Levels of activities may be very different depending on the level of activity and on the objective of measurement, online (quality control or safety alarm) or afterward.

Tritium disintegration produces an electron, an atom of ^3He and liberates 325 $mW.g^{-1}$.
These three types of information are used for measurement. It depends on the physical form and level of activity.

For gas releases, electrons are measured in an ionisation chamber as an electrical current (>40 Bq/L for a 1L chamber). This current is also used and amplified in a proportional counter which needs a counting gas but delivers a better information (>3 $Bq.L^{-1}$ 1.3 L and 1 mn). The chemical form (HT, HTO) is not accessible by these methods. At industrial scale, outgassing of drums is measured above 0.5 $MBq.d^{-1}$.
The liquid scintillation technique is also often used to measure gas, after transformation into liquid. To obtain water, air is sampled continuously and sent for bubbling in a set of two or more flasks of pure water with a given flow rate. Tritium in the HTO form is so collected. Remaining tritium (mainly HT) is then oxidised (thermal or chemical technique) and sent through a second set of flasks. The water is then analyzed after a time of charge. Tritiated water is added to a scintillating cocktail which transforms β (e-) in photons. Photons can then be collected and measured out of the liquid. This technique has the advantage to measure separately HT and HTO. Nevertheless, it may be very sensitive to artefact like other radioactive gas (^{14}C, Rn), chemiluminescence, static electricity. It is easy to measure 10 $Bq.L^{-1}$. It is always possible to dilute if necessary for high activities. It may be useful to distillate water in the sample preparation.

Gas inventory can be obtained by calorimetry or microcalorimetry, with a measurement of heat flux (energy liberated) in an isotherm calorimeter. Detection level is about10 $nW.mL^{-1}$ of tritium. Another possibility is the solid scintillation technique, using a crystal which transforms the electronic excitation in a photon emission.

The last possibility is the measurement of ^{3}He. It is used in Valduc for activity of drums, the detection limit is about few GBq/drum at industrial scale. This technique is also used for measurement of very low level of activities in water. Values as low as 0.01 $Bq.L^{-1}$ can be measured by spectrometry measuring ^{3}He after many month of delay.

Mass spectrometry is used to quantify isotopes and impurities. Molecules are introduced in a source of ions. These ions are accelerated, filtered in a mass spectrometer (trajectories depend on the ions mass on charge ratio) and collected in a Faraday cage.

Micro gas phase chromatography with a cryogenic column is also a more recent method to distinguish the different species of a gas based on their permeation characteristics.

The above two techniques can be coupled.

Raman spectrometry is based on the measurement of the frequency of diffused light when a monochromatic light source interacts with molecules (more than monoatomic). This technique enables qualitative and quantitative chemical analysis on tritiated gases, even for mixed molecules (HT, DT and HD). The task is to determine 0.5 % HD in 99.5 % ($H_2 + D_2$).

Integrated in glove box, ICP-AES can determine metallic impurities dissolved in tritiated waters with the aim of choosing the correct way of treatment for these waters. This technique enables the precise quantification of values as low as 0.1 $mg.L^{-1}$ impurities (Fe, Ni, Cr, Al, S…) in waters.

5.4 Tritium transfer in environment

Hydrogen is a major element and water is the regulator medium for life on earth. A lot of mechanisms exist to maintain life in the fluctuation of its environment.
For this reason, models for hydrogen and water belong to two categories: one dealing with routine releases all over the year, with numerous but very simple relations between compartments; the other dealing with accidental releases where it is necessary to get a lot of

information and elaborate models. We shall develop separately these two cases starting with the pseudo-equilibrium approach for routine release.

5.4.1 Tritium transfer for normal release

The full approach for normal release is very well described in the reviewed Tecdoc – and TRS 364 of the IAEA to be published in 2008.

The simplest approach was in the past to consider water vapour of air at a given point and to suppose that any water in soil, plant, animals was in equilibrium. The dose calculation is so limited to the average concentration in vapour multiplied by a 1.3 $kg.d^{-1}$ rate of consumption and by the dose per unit intake factor. This approach gives a good order of magnitude but nowadays does not appear reliable enough.
Ingestion is the main exposure pathway.

According to traditional usage, the "Specific Activity" model has been used for tritium, considering that the ratio $^3H/^1H$ is kept from one compartment to another one.

For tritiated water, the "specific activity" model is formulated in terms of tritium concentrations in water rather than the ratio of tritium activity to the mass of hydrogen in a given compartment.
Measurements give masses of dry matter, free, and combustion water, and concentration in the different waters, corresponding to three different types of tritium: free water for tritiated water, exchangeable organically bound tritium (OBT) which can be extracted when repeatedly washed with tritium-free water, and non-exchangeable organically bound tritium which remains at the end.
This approach needs to define a water equivalent factor for organic mater that can be deduced from content of hydrogen in protein (7%), fat (12%) and carbohydrate (6.2%). For most of the plants this value is about 0.5 – 0.6.
The second important point is that because of the difference of mass, there is a discrimination factor between ^1H and ^3H in the different transfers and biochemical transformations. From measurements, this factor is near 0.5.
In practice, it is still possible to have a simple assessment more realistic, with the following equation:

$$C^{total}_{plant.fw} = [H_{rair} + 0.3(1 - H_{rair})].\left[(1 - H_{plant}).W_{eq}.D_p + H_{plant}\right]C_{air.w}$$

Where H_{rair} is the relative humidity of air, H_{plant} the proportion of water in plant, W_{eq} the water equivalent of dry matter, D_p the discrimination factor between ^3H and ^1H.

As an example, considering a relative humidity of air of 70%, a proportion of water in green vegetable of 80% and in wheat of 20%, a W_{eq} of 0.6 and a D_p of 0.5, the relation would give respectively the activity of green vegetable equal to 2/3 of the air vapor concentration and the activity of wheat equal to 1/3. OBT represents 7% of the tritium for green vegetable and 54% in the case of wheat. This means that compared to the rough model, the activity of vegetable would be about a factor 2 below. This approach has the advantage to be more demonstrative, separates organic compounds and water and finally gives average value that can be compared to measurement.

For tritium gas (HT), the dry deposition on ground is about 10 times below HTO dry deposition and there is neither deposition by rain nor direct transfer from air to leaves. Dry deposition is quickly transformed in soil in the HTO form by micro organisms. Then HTO enters the previously described cycle of tritiated water. From these bases confirmed by field measurement, HT impact is about few % of HTO impact for a same activity release.

In reality, for survey purpose, there are fluctuations around these average values in the different compartments of the environment depending on the wind direction fluctuations, the source term fluctuations and the residence period of water in the compartments. Free tritiated water has period of few hours in plant. On the contrary, non exchangeable OBT behaves as an integrator for the period of growth of the sample measured. Soil needs many months to renew its water. These elements weighted in the case of a more or less continuous release are much more important in the case of an accidental release.

5.4.2 Tritium transfer for accidental release

In case of an accident, the first exposure comes from inhalation and transcutaneous contamination from the contaminated cloud. Then the contamination of plants by exposure to HTO in air results in doses during the first few days after the release, while the OBT produced by air contamination remains in the plants up to the harvest, with the first crop delivering the main exposure. Finally, contamination via the soil delivers the remaining exposure till the following winter when leaching decontaminates the soil. Some edible parts like potatoes or cereals grains are stored. It delays their dose impact for many months.

Mechanisms of concern for HTO are: incorporation rate from air to leaves, dry deposition and rain deposition from air to soil, transformation of HTO to OBT into the leaves, translocation from leaves to fruits, roots and tuber, and at last the turn-over of water in soil.
All these mechanisms have been modelized at different extension and there is still a need to synthesize the different mathematical approaches and to harmonize and/or complete knowledge as it can be seen in the following paragraphs.
Within the IAEA program EMRAS, an analysis of the consequences of an acute atmospheric release of tritium as been performed [EMRASS 2008] for a 10g accidental release occurring in the mid-June, when plants are growing at their maximum rate. Three types of standard meteorological conditions have been evaluated : case 1 = **sunny day** with unstable conditions and 2 $m.s^{-1}$ wind – case 2 = **rain day** with neutral conditions, 5 $m.s^{-1}$ wind and 15 mm rain – case 3 = **clear night** with stable conditions, 2 $m.s^{-1}$ wind.

Considering all pathways together, Figure 4 shows that there is great variability of results that would lead, depending on the assessment, to useful counter measures or no interventions [Guétat 2008].

As in a previous work [Davis 1995][Galeriu 2008], the distribution of results varies depending on the scenarios and model approaches. The differences for rain and night cases reach one order of magnitude; this is in part due to atmospheric dispersion models but mainly to the modelling of the food contamination.

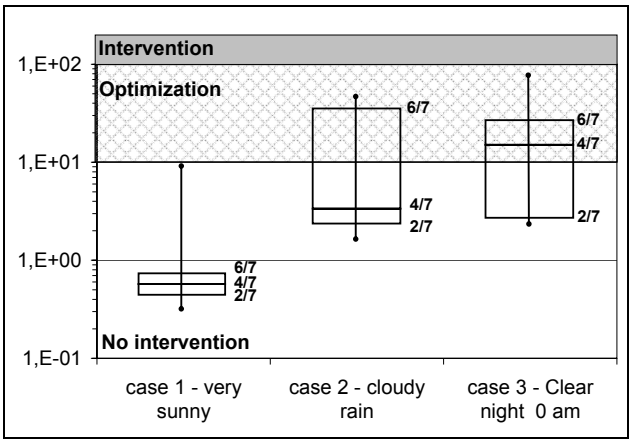

Fig. 4: Distribution of total dose for a 10g tritium release under three different meteorological conditions at 1 km.

Considering the three cases, the fact that OBT production is less important during the night is obscured by the differences among atmospheric dispersions.

To suppress uncertainties resulting of dispersion models, effective dose results (Fig. 5) are compared after normalizing to a mean air concentration.
It is clear that food is the main contributor to dose and that ingestion of cereal and vegetable represents a large part of the total dose, the relative importance of each depending on the model used. Dose impact due to ingestion of cereals is high because tritium is in the form of OBT, and cereals can be stored for a long time. However, it is unlikely that the entire daily consumption of cereals (430g.d^{-1}) is composed of highest level contaminated wheat coming from a single location because of dilution within the harvest industrial process. Assuming all cereals come from one location, models (pessimistically) predicted that cereals contribute to 10% to about 80% of the exposure. Cereals are clearly the main source of uncertainty (overestimation) in these exposure assessments.

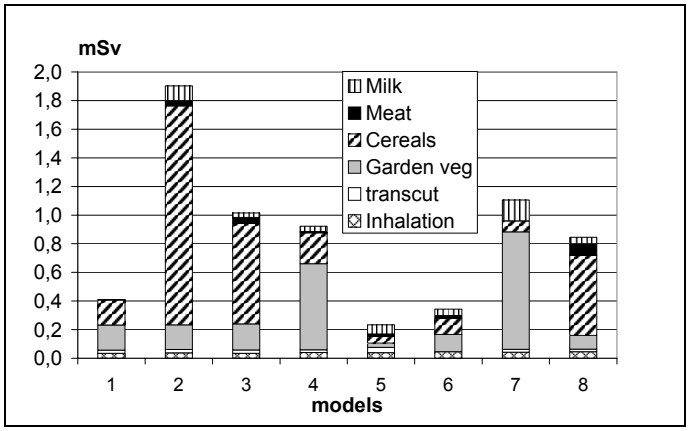

Fig. 5 : Effective dose details by pathways (normalized air concentration)

In the case of tritium, climatic conditions are not limited to atmospheric dispersion conditions, however it is necessary to include all parameters having influence on the photosynthesis mechanisms and agricultural parameters (crops characteristics and cultural techniques). Fig.6 and 7 present results cited in [Barry 1999] to illustrate the influence of the precise hour and date of accident.

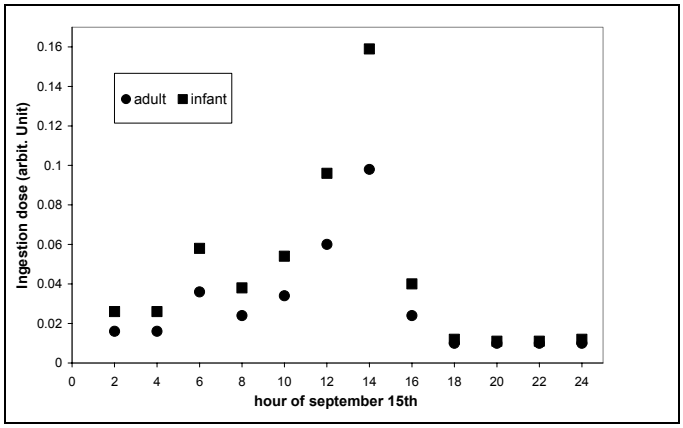

Fig. 6: Exposure versus hour of accident on a given day of September (arbitrary unit).

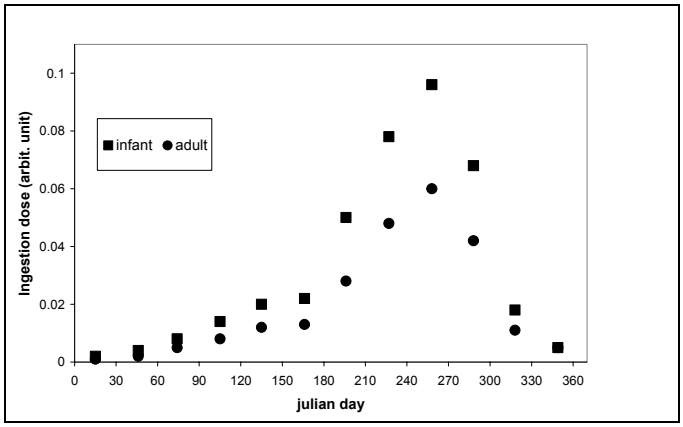

Fig. 7 :Exposure versus date of accident in the year.

From these two theoretical documents, it is clear that we don't have presently enough certitude on acute release assessment.
Depending on the exercises, it is not evident that rain has or not a strong effect on total exposure. In the Emrass exercise, the answer is negative, but in other documents, increases of deposition up to a factor 7 are shown.

In the EMRASS exercise, for most of the models, OBT elaborated during air contamination is the main exposure source. Air pathway is slightly dominant but the difference between air and soil pathways is generally not very high. Nevertheless big differences exist for the soil water contribution which depends on two parameters, deposition velocity and turnover of water in the soil. It is confirmed that a tritium release has an impact via soil contamination for some months (one agricultural season) and that the exposure has so to be integrated on this duration.

This exercise also shows that there is a need for industrial production like cereals to calculate average activity at the scale of a field (and not at a point) and be precise about the nature of consumption product and dry matter content. Over estimation by an order of magnitude may result of such misunderstanding of data.

Model predictions are still highly uncertain and differences between models are high. In particular not enough is known about OBT produced at night, about what the dynamics of soil deposition and movement in soil are, and finally what the fraction of HTO in the leaves coming from soil is. This can be shown on three participant results whose models could give activity values at different times (Fig 8).

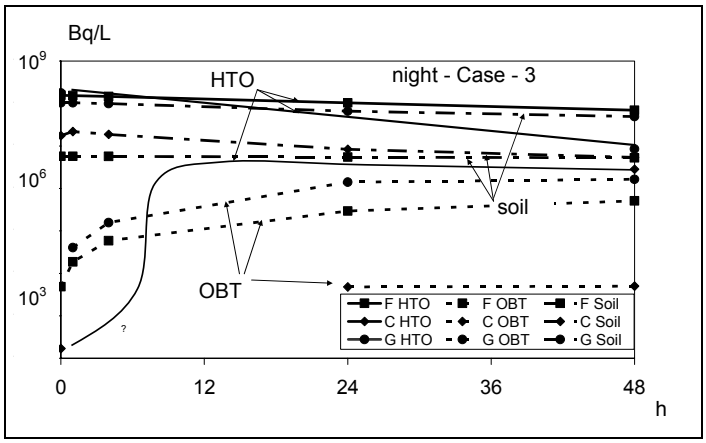

Fig. 8: Tritium concentration during the 1st two days in the case of a midnight contamination, calculated according to three models.

Another EMRASS exercise dealt with experimental work on sojabean contamination. The incorporation rate of HTO in OBT at different times between flower and harvest is shown in Figure 9. Variations by a factor of three orders of magnitude are shown. These data are plant dependant. There is a need to obtain or use agronomical data of this type to increase the accuracies of assessment.

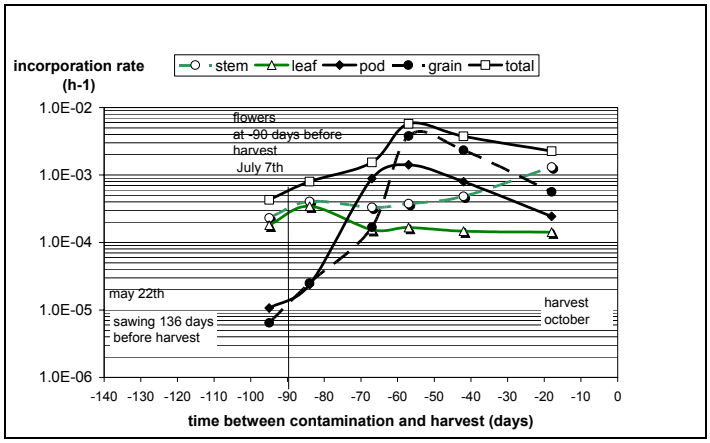

Fig. 9: OBT incorporation at harvest versus time of contamination.

Some other interesting information comes out:

- For the young children, without intervention, the total dose does not appear significantly different from the adult total dose.
- Activities of animal products are lower than vegetable activities.

For a HT release, the impact is about 1% to 3% of an equivalent release of activity in HTO. Here again, this can be explained by a lack of air-plant contamination (only reemission taking account of the wind rose) and a smaller deposition velocity (about a factor of 20) [Davis 1995].

5.4.3 Regulatory aspects for tritium

Tritium is a β emitter with a very low radio toxicity and an average energy per disintegration of 5.7 keV (to compare with other major βγ emitters having energy of about 1 MeV.).. However tritium can be relatively easily measured.
In most technical documents based on radiological impact, limits for tritium are much higher than for other usual radionuclides. This is the case for the European directive on radiation protection, and the World Health Organisation norm for drinkable water [WHO 2004].
But quite often, regulatory values refer to group of nuclides or type of emission - βγ emitters - In this type of document, tritium is generally considered as other nuclides and so gets values much lower: it is generally penalized by one and often two orders of magnitude.
This is the case for the surface contamination in transport regulation, and could be the case for limit for food trade after accident. We shall develop here this last point as it has not been already taken into account in a law.

The dose level of 10 mSv in one year for people living in self sufficiency has been considered by the International Commission of Radiation Protection [ICRP 1991]. The Codex Alimentarius Commission considers a dose of 1mSv for a year after an accident, 650kg.y^{-1} of consumption and 10% of food from local origin [ALINORM 2006-04]. This commission has also adopted [ALINORM 2006-07] a value for trade of 10^3 Bq.kg^{-1} for infant foods and 10^4 Bq.kg^{-1} for other foods [ALINORM 2006-04]. It must be noted that corresponding doses are evaluated respectively at 2 µSv and 20 µSv, and not 1 mSv like for the other elements.

In the EMRASS scenario, the modellers calculate concentrations in vegetables at 1h and 48h with the Codex dose and consumption hypotheses also consistent with ICRP approach (Fig 10). Two predictions have to be excluded, one in the night case for one model that does not evaluate the incorporation of tritium during the night, but only considers the soil contamination at the following morning; the other does not include HTO coming from the soil during a sunny day after 48h.
All models predict concentration between 5 10^6 and 5 10^9 Bq.kg^{-1} one hour after accident and between 2 10^6 and 1 10^8 Bq.kg^{-1} at 48 h for fresh leafy vegetable.
These values are independent on the weather conditions that occurred during the release. These values may be compared to USDOE guidance 10^7 Bq.kg^{-1} [US DOE 2006], or the action levels from Canadian guidelines (3 10^4Bq.kg^{-1} for milk and 10^5 Bq.kg^{-1} in OBT form for commercial foods [18].

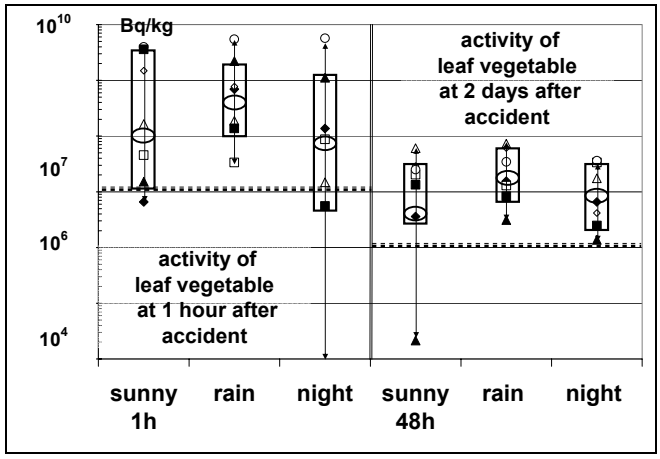

Fig. 10: Activities of vegetable at 1h and 48h after the accident corresponding to exposure criteria used to establish food trade limits.

5.5 CONCLUSIONS

5.5.1 Concerning the sources of tritium, some conclusions can be drawn

Natural tritium production is higher than man-made release.
Atmospheric nuclear tests have multiplied by 100 inventory and concentrations at world scale and even more. The effect on atmosphere is now finished.
Reprocessing plants and HWR are the main sources of releases, reprocessing plants releasing mainly to sea in the EU.
Heavy water reactors produce much more Tritium (D-T) than PWR, and PWR (boron) more than BWR (ternary fission). T production of reactors increases.
Some specific uses generate specific impact (dissolved organic products, lighting devices).
Waste inventory is very small, but inappropriate limits for tritium in water table generates a need of interim storage.
Fusion will increase the use of tritium but should not change fundamentally release level. Treatment conditioning of waste could be a significant source.

5.5.2 Concerning measurement, the conclusions are the following

Measurement apparatus for large inventories and large volumes exist but are to be improved.
Generally, it is easy to measure tritium at a low level but that needs to have a good preparation.
Sample preparation takes time for solid material, and it would not be easy to manage in case of incident.
For wastes, it is not easy to reach ANDRA's surface disposal requirements.

5.5.3 Concerning Transfer in the environment, the conclusions for normal releases are the following

For normal tritium release assessment, IAEA document TRS 364 –to be published in 2008 and the corresponding TECDOC– will constitute an up-to-date synthesis.
For HTO: At each step of the transfer chain, dilution occurs; Vegetable Ingestion is clearly dominant and Direct Air-plant pathway is dominant.
For HT: release has a 1-10 % dose impact compared with HTO, and exposure comes then from HTO converted in soil.

Tritiated water of plant gives information about the few last hours; OBT at the contrary is an integrator.

In the case of accident or incident:
From the scientific point of view, general features of tritium behaviour are known. Tritium does not concentrate in food chain, but can remain in soil for an agricultural season.
The variability remains very large in case of accident especially in rain and night cases. On the contrary, the relationship between dose and tritium concentration in the plant water during the first two days is not really influenced by meteorological conditions and scenarii considered: this emphasizes the interest and necessity of sampling after an accident.

Modelling demonstrates, however, that between 1h and 48h, the concentration of tritium decreases and the OBT/HTO ratio increases. It is thus important when defining the reference values for intervention to take into account these changing concentrations, the chemical form of the tritium and the type of vegetable.

Although a release of 10 g of tritium (HTO) has a small radiological impact. Some limited intervention close to the release point may be necessary depending on meteorological and environmental conditions.

A release of 1kg of Tritium (HT) has the same radiological impact than 10g of tritium (HTO), but the main pathway is the soil contamination.

Experiments are needed to reduce the uncertainty concerning wet and dry deposition velocity onto soils, the uptake by plants of HTO from air and soil during the night and the translocation of OBT throughout the plant.

The present “tritium scientific community” is small and has to synthesize what is absolutely needed in models. This community could disappear from EU in the few next years.

5.5.4 From the regulatory point of view

Tritium has to be considered apart from the other beta-gamma elements because of its low radio toxicity, and because of the difficulty to confine it.

The limitation at 4 $Bq.cm^{-2}$ for beta-gamma in transport regulation should be adapted for tritium.

The concentrations in foodstuffs recommended by the Codex Alimentarius as intervention levels are much too low for tritium, with its low-level beta radiation: the tritium doses resulting from these concentrations are much lower than the $1mSv.y^{-1}$ criteria.

For drinkable water, WHO value (10^4 $Bq.L^{-1}$) should be kept as a reference. Moreover 100 $Bq.L^{-1}$ should be kept as an investigation level but not understood as the drinkable limit. This would be very useful for surface disposals.

5.6 Acknowledgements

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5.7 References

ALINORM (06/29/12) - Codex Alimentarius Commission report on the 38th session of the Codex Committee on food additive and contaminant The Hague Netherlands 24-28 April 2006.

ALINORM (06/29/41) Codex Alimentarius Commission 29th session Geneva Switzerland 3-7 July 2006.

Batifol G., Sejournant C., Douche (2008) C. - Tritium release reduction and radiolysis gas formation - TRITIUM 2007 “8th international conference on tritium sciences and technology Rochester” - *Fusion Science and Technology* 54 n°1 ; 215-218.

Barry P.J., Watkins B.M. , Belot Y. Davis P.A., Edlund O. Galeriu D., Raskob W., Russell S. Togawa O. (1999) - Intercomparison of model predictions of tritium concentrations in soil and foods following acute airborne HTO exposure. *Journal of environmental Radioactivity 42 (1999) 191-207*.

Carmin (2005) - Comte A. CEA Cellule Carmin – Tritium, Fiche radiotoxicologique.

Davis, P.A., Galeriu, D (1995) - Evolution of HTO concentrations in soil, vegetation and air during experimental chronic HT release V Topical Meeting on Tritium Technology in Fission, Fusion and Isotope Applications, Italy, May 1995. *Fusion Technology* Vol.28; 833-838

Galeriu D, Davis P, Chouhan S, Raskob W. (1995) - Uncertainty and sensitivity analysis for the environmental tritium code UFOTRI. *Fusion technology* Vol.28; 853-858.

Galeriu D., Davis Ph., Raskob W., Melintescu A. (2008) - Tritium radioecology and dosimetry – Today and tomorrow - TRITIUM 2007 “8th international conference on tritium sciences and technology Rochester” - *Fusion Science and Technology* Vol.54 n°1; 237-242.

Guétat Ph Patryl L. (2008) - "Environmental and Radiological Impact of Accidental Tritium Release"- Tritium 2007 “8th international conference on tritium sciences and technology Rochester”, *Fusion Science and Technology* Vol.54 n°1 ; 273-276.

HEALTH CANADA (2007) - Canadian Guidelines for the Restriction of Radioactively Contaminated Food and Water Following a Nuclear Emergency.

IAEA EMRASS - hypothetical scenario to be published - Draft on http://www-ns.iaea.org/ projects/emras/emras-tritium-wg.htm.

ICRP (1991) - publication n° 63, *Ann ICRP 22 Pergamon Press, oxford*.

Jones G.C. (2008), PWR Tritium Issues TRITIUM 2007 “8th international conference on tritium sciences and technology Rochester“, *Fusion Science and Technology* 54, to be published

Mihok (2008) – Tritium in precipitation in the vicinity of stacks at Canadian nuclear facilities - “TRITIUM 2007“ “8th international conference on tritium sciences and technology Rochester“, *Fusion Science and Technology* Vol.54, to be published

Mutch R D., Mahony J (2008) - A Study of Tritium in Municipal Solid Waste Leachate and Gas “ TRITIUM 2007" “8th international conference on tritium sciences and technology Rochester“, *Fusion Science and Technology* Vol.54 n°1 ; 305-310.

UNSCEAR (2000). Sources and effects of ionizing radiation. Report to the general assembly with scientific annexes. Volume 1.

US DOE (2006) - United States Department of Energy - Memorandum: Guidance on Deriving Intervention Levels for Tritium Contaminated Crops and Animal Feed for DOE Emergency Planning and Response Activities, may 2006.

Wallis M. K. (2005) - UK regulatory Performance in practice – the Amersham Cardiff nuclear site. *7th SRP international symposium (*12-17 June 2005).

WHO (2004), World Health Organisation - Directives de qualité pour l'eau de boisson. Vol.1. Troisième édition. Genève, Suisse.
http://www.who.int/water_sanitation_health/dwq/gdwq3rev/fr/index.html

6 PLASMA WALL INTERACTION IN FUSION MACHINE (MICRO-PARTICLES CREATION PROCESSES)

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6.1 Introduction

In physics, nuclear fusion is the process by which multiple atomic particles join together to form heavier nucleus. It is accompanied by the release of energy. Fusion reactions power the stars. It takes considerable energy to force nuclei to fuse, even those of the lightest element like hydrogen. This is because all nuclei have a positive charge (due to their protons), and as like charges repel, nuclei strongly resist being put too close together. Accelerated to high speeds that is, heated to thermonuclear temperatures, they can overcome this electromagnetic repulsion and get close enough for achieving fusion. The fusion of lighter nuclei, creating a heavier nucleus and a free neutron will generally release more energy than it took to force them together. This is the case of the fusion reaction commonly used in fusion reactors where a mixture of Deuterium and Tritium is heated to more than 200 Billion degrees. At these temperatures, fusion is observed and helium nucleus is created with a high energy neutron. This reaction releases a huge amount of energy.

At this very high energy, plasma is obtained which is an ionized gas where the electrons are separate from the nucleus. It is the fourth state of matter. Plasma is a mixture of ions and electrons. In order to achieve fusion reactions, ions and electrons must be confined as well as possible in a closed volume. In a star, this is achieved by gravitational confinement. On the earth, confinement is performed by high magnetic field obtained by superconducting coils. The fusion machine which has proven to be the most efficient in producing fusion reaction is the Tokamak. It is like a giant donut. The centre of the donut is occupied by the plasma surrounded by the magnetic coils. In a Tokamak, before any fusion experiment, the inner volume of tens of m^3 is first completely emptied of all occupying gases. Then, plasma fuel (50% of deuterium + 50% of Tritium) is injected at a very low pressure (10^{-4} atmospheric pressure) and heated to high temperature in order to realized fusion reactions.

Magnetic confinement is the most efficient confinement on earth. However, this magnetic confinement is not perfect and particles escape from the confined media and interact with the first material wall they find on their trajectory. Even if the temperature of the ions impinging the surface is not at the same huge level than in the centre of the plasma, the interaction of these plasma outflows with plasma facing components leads to severe constraints on the material. These issues are described in the following chapter.

6.2 Plasma wall interaction in Tokamaks and Tokamak tritium cycle

In order to sustain in steady state such as high heat outflow escaping from the plasma, it has been necessary to develop new plasma facing components (PFCs) technology able to

survive to this constraint. These new developments are based on actively cooled components which can sustain up to 20MW/m^2. It has to be recall that this power is comparable to the energy observed at the Sun surface. These new PFCs were first tested in Tore Supra (the French Tokamak) several years ago and permit to this machine to establish a world record of extracted energy in steady state (more than 1 Mega Joule) and the realisation of the longer plasma pulse never seen (more than 400s). Further developments successfully tried to up to 30MW/m^2 in heat flux simulator, have shown that this technology is mature to be used in fusion machine like ITER. This great success consolidates all the fusion actors in the ITER PFCs solutions.

One consequence of this plasma outflow interaction with wall is the sputtering of the materials which form the PFCs. This erosion leads to re-deposition of material. In this process part of the fuel of the discharge is trapped in the deposited layers. From current assessment, it is considered that part of the injected fuel could be trapped in this layers leading to an in vessel storage of tritium of up to 5g of Tritium per ITER shot at fuel power.

6.3 Dust creation processes and ITER perdictions

As we have already seen, during Tokamak operation and due to high heat and particle fluxes, Plasma Facing Components (PFCs) are eroded and material is re-deposited. Mainly due to internal stresses, these re-deposited layers are fragile and breakable leading to micro particles creation. For ITER extrapolations, it is of major interest to know the proportion of these deposited materials able to be converted in airborne dust. In order to quantify this transfer process, a dust conversion factor (Cd) can be used. Cd is the ratio between the total quantity of in vessel dust (Qd) that can be recovered at the opening of the machine over the total quantity of eroded material (Qe) produced during all operation processes:

$$Cd=Qd / Qe$$

The estimation of Cd is based on the measurements of all the airborne dust collected and on the estimation of the eroded material during all the Tokamak erosion processes. In this Cd evaluation, all the eroded materials are supposed to be deposited and then part of them turns to be converted in dust. The upper limit of the in vessel dust quantity at any time could be obtained knowing the eroded quantity (thus, Cd=1). However, this constraint could be released with a reduced Cd.
Cd was assessed for a 5 months (1438 shots) Tore Supra operation period. The total in vessel dust collected (with a vacuum cleaning system) during this campaign was 31g. In order to evaluate global Tokamak erosion, all the erosion processes have to be reviewed and eroded quantity assessed:

- Erosion during normal operation. The eroded quantity results to the sputtering of the carbon material by the plasma out-flux which is equal to $Np/\tau p$ with Np being the plasma particle mean density and τp, the characteristic residence time of particles in the plasma. Considering a carbon sputtering yield equal to 0.02, C eroded quantity is 27g. However, this estimation is an upper value of the eroded carbon during normal operation. Indeed, it has been proved that 50 to 80% of the eroded carbon is re-deposited locally and takes part afterwards to the ongoing erosion process. As a consequence, the erode value could be as low as 27*0.2 ~5.4g.
- Erosion during disruption which is a lost of plasma confinement event in normal operation which leads to the release of the stored plasma energy in less than 20ms.
  The contribution of disruptions is much more difficult to assess. The thermal content of the discharge is known (~300kJ) but the surface of the interacting zone is not addressed precisely. However, from experiments results and code evaluation presented by

Hassanein, it appears that the eroded quantity (Mc) per disruption could be approximated simply by:

$$Mc\ (g) = [(Eth * 0.1) / Esub] * 12\ (g)$$

Where Eth is the plasma thermal energy before the disruption and Esub, the C sublimation enthalpy. Here, it is considered that the time duration of the disruption is so small that Eth is deposited on the PFCs surface and contributes to sublimation. With the Tore Supra operating parameter, the eroded material per disruption Mc is 0.6g. Considering a disruption frequency of 10-15% during the considered period, this leads to 80 to 120 g of PFCs erosion.

- Erosion during maintenance activities like conditioning, normal procedure to prepare the in vessel material for the vacuum operation. Helium Glow Discharges are used in Tore Supra. Glow Discharge are low temperature, low density plasma.
  During Tore Supra (He-GD) (3A of Glow current, 300V glow voltage), the net erosion is $1.8x\ 10^{18}$ Carbon/s, considering a carbon sputtering Yield of $9x10^{-2}$ at 300eV. For 1 day of He-GD, 3g of C are sputtered. During the 5 months of TS operation and with regular He-GD cleaning sequence, at least 50g of C could have been eroded. With the Tore Supra discharges parameters used, no dust production is observed and the eroded material is re-deposited as layers.

Several observations have shown the influence of moisture on the evolution of carbon deposited layers as a function of time. Cracks appear leading to embrittlement of the deposited layers, flaking and thus to dust creation. However, it seems that it takes days to observe this layer destruction in Tokamak at room temperature. This flaking process has not to be included in this study since micro particles are recovered just after the opening of the machine.
The quantity of Carbon eroded in Tore Supra during this 5 months operation campaign is therefore composed of:

- 27g eroded during normal operation (this is an upper value that could be reduced by a factor 5)
- 50g eroded during HeGD
- 80-120g eroded during disruption.

Total eroded quantity is close to 200g, the main erosion pathway being clearly disruptions.
For this Tore Supra campaign, Cd is equal to 15% which corresponds to the ITER retained value. The value recently published by JT60U is comparable: 7%. From ITER calculation and in the frame of the current design, 50g of material are eroded per shot leading to a dust production of 7.5g.
For dust inventory reasons as well as for Cd evaluation, the ITER in vessel quantity of airborne dust (Qd) must be assessed. In the following chapter, the in vessel diagnostics able to weigh up Qd and currently available are reviewed.

6.4 Current dust sample characterisation

Dust have been collected and characterised in a number of fusion devices around the world. More than 10 devices where involved. The collection system used was the same for all the devices: the surfaces were scratched with a brush and the material obtained captured by filtered vacuum collection. Then several analysis were undertaken like sizing and counting (with optical microscopy imaging), BET measurements in order to assess the Specific Surface Area and or the dust shaping evaluation as well dust composition including Tritium when Tritium was used as a fuel.

Even if a dispersion of the data was observed, it is possible to extract general tendencies about the physical and chemical properties of these micro particles.
The particle Count Median Diameter is 2.8 +/- 2.4 µm. However, the size of dust particles in fusion devices is rather large from 100nm to more that 10µm.
The shape observed is very complicated in term of topology. Pure spherical particles are observed. However, fractal shape as well as nano tubes or fibres are also observed in the samples obtained.
Several studies have been undertaken with tritiated dust in order to assess the Activity Median Aerodynamic Diameter. In JET, the EU Tokamak able to operate with Tritium, airborne dust were collected by membrane filter air sampling and with a cascade impactor device. In the case of the membrane filter, AMAD was estimated to be ranging between 1 to 7µm. In the case of the impactor, multi modal distribution were observed between 1 and 20µm, the later being the most abundant. This value is comparable to those already measured in TFTR, a US tokamak operating with Tritium in the 90s.
The Surface Specific area is in between 1 and 20 m^2/g.
The composition of the dust is nominally shares the composition of walls that interact with the plasma. However, some discrepancy can be observed in the same machine. In Tore Supra, pure carbon micro particle are observed as well as mixture of Carbon and metal material. This is coming from the different operating plasma scenario of the machine.
Finally, it is obvious that the majority of dust particle in Tokamak is found in the bottom of the machine. However, some part of the dust recovered are also found in duct and in between the PFCs of the machine.

6.5 Dust in vessel measurements and control

6.5.1 In vessel micro-particles measurements

In order to assess the Cd conversion factor, PFC erosion diagnostics are needed. The most common technique used in current tokamak to assess Qe is code evaluation of the erosion knowing the plasma edge impurity diagnostics. This was used in Tore Supra and JET. However, this technique is well suited for in vessel wall erosion assessment but much more difficult for divertor area due to difficulty to diagnose the impurity in the divertor volume. For a more reliable measurement, gross erosion techniques able to measure the PFCs depth evolution as speckle interferometry or laser metrology could be used. These techniques, developed at the laboratory scale, must be integrated and tested in Fusion machine environment. The capability of deducing global measurements from local ones due to reduce measurement zone must be also addressed. It has to be stressed that erosion diagnostic is the only one that could give the envelope value of the in vessel dust quantity considering Cd=1. In that sense, it can be regarded as the only diagnostic that could be able to track dust inventory evolution as an upper value.
For airborne dust, several diagnostics operating on a shot to shot basis are under development. Electrostatic grids could be installed in places where dusts are accumulating as under the divertor or under the ITER dome. This system relies again on homogeneity assessment and the link with local measurements and Qd is not obvious. Optical techniques could be also used. Laser extinction is the simplest one since it is measuring the attenuation of the laser light intensity along its propagation in the dust cloud. However, the interpretation of the measured signal turns to be very difficult due to the heterogeneity (in size, composition, shape,…) of the micro particles supposed to be produced during the ITER operation. Furthermore, this airborne dust measurement relies on the interaction of gas and PFCs which is supposed to put the micro particles in suspension as it occurs during an air in vessel ingress. The link between the airborne dust measured and the dust really mobilisable relies on complicated fluid codes that do not exist yet. However, optical system as laser

extinction could be inserted on in vessel inspection robot and thus available for global assessment independently of in vessel in-homogeneity.
Finally, airborne dust is simply measured via Vacuum Cleaner recovery that can be introduced remotely and used during very short shutdown. This diagnostic system that could be also used for dust removal has already proven it efficiency. It seems nevertheless that accessibility will be an issue as well as the link between dust recovered and the real quantity of dust that could be mobilised.
As for erosion the need of Tokamak (or scale one mock-up) integration and test is mandatory in order to assess these diagnostics capabilities and accuracy in a realistic condition
As a conclusion, a set of system seems to be available to measure the in vessel airborne dust. This value could be then compared with the given today ITER administrative guidelines for limiting in vessel dust inventory. Approaching to these values; removal tools must be used.

6.5.2 In vessel micro-particles removal techniques

Micro-particles in vessel removals rely on rather clear procedure. It is first necessary to unstuck the particles from the surface. Due to Van Der Walls, the action force needed is inversely proportional to the size of the particle. Then particles are collected and removed from the vacuum vessel. Two techniques could be used in order to mobilise the dust: high pressure gas injection and laser matter interaction.
Gas has been proposed but it has never been tested with realistic micro-particles and in tokamak topology. The gas inventory control and treatments must be part of the assessment of this collection technique and has also to be addressed. When the particles are unstuck and mobilised, Vacuum Cleaning suction is the easiest one and already tested in JET.
The laser mobilisation technique could be useful to access rather small structure like castellation and for small particle collection. Experiments with laser have already been done and have demonstrated the high laser efficiency for dust in castellation suspension.
Laser interaction give birth, under vacuum, to high speed small particles and collection by sticking on retrievable embarked system could be preferable in order to avoid huge amount of gas collection and treatment. Dusts are rapidly negatively charged in a low Temperature, low density plasma like Glow discharge. Therefore, laser dust in glow plasma could also be used for micro-particles collection. This technique could be then easily upgraded to tritiated dust collection since these micro-particles will be naturally charged due to Tritium beta disintegration. At higher pressure, vacuum cleaning suction could be also possible as for gas injection.
Several others proposals as liquid vacuum vessel washing have also been proposed. This rather complicated technique must be tested in mock-up prior to any test in the severe tokamak environment.
To conclude, the dust removal procedure relies on already checked systems. However and as for the dust diagnostics, Tokamak integration and test must be foreseen as soon as possible to check the reliability of the techniques proposed in a realistic environment. However, it is conceivable that a set a technique will be used in order to achieve the ITER goals.

6.6 Conclusions

In this report, the 2 major consequences of Plasma Wall Interaction with Plasma Facing Components in Tokamak were presented. It has been shown that the technological development of actively cooled components allows the Tokamak high energy steady state operation. This technique is now ready to be used in ITER. In parallel, Erosion of the PFCs leads to material redeposition and then to dust creation due to layer cracking.

The dust conversion factor has been estimated in a current operating Tokamak. It is of the order of 10% which is close to the value retained in the ITER dust production evaluation. Using the procedure presented here, Cd is under estimation among several operating worldwide Tokamaks in order to address the consequences of different operating mode (influence of ELMs for example) and material configurations (metal and non metal) on the dust in-vessel quantity.
The dust was recovered from several machine and the dust properties reported. The AMAD is shown to be equal to 7µm.
Finally, several diagnostics and removal system have been presented above. It appears clearly that a set of techniques are available to assess and control the dust in-vessel inventory. However, there is an urgent need of an integrated demonstration that has to be planned in a Tokamak environment or in a scale one to ITER mock up.

6.7 References

J.P. Sharpe, D.A. Petti, and H.W. Bartels, Fusion Engineering and Design 63-64, (2002) 153-163.

Ph. Chappuis, E. Tsitrone, M. Mayne, X. Armand, H. Linke, H. Bolt, D. Petti and J.P. Sharpe, J. Nucl. Mater., 290-293, (2001), 245.

J.T. Hogan, E. Dufour, P. Monier-Garbet, C. Lowry, Y. Corre, J. Gunn, R. Mitteau, E. Tsitrone, C. Brosset, J. Bucalossi, T. Loarer, I. Nanobashvili, B. Pegourie, P. Thomas, Proceedings of the 21st IAEA Fusion Energy Conference (2006)

A. Hassanein, V. Belan, I. Konkashbaev, L. Nikandrov, V. Safronov, A. Zhitlukhin and V. Litunovsky, J. Nucl. Mater., Volumes 241-243, (1997) 288.

E. Salançon, T. Dürbeck, T. Schwarz-Selinger and W. Jacob, J. Nucl. Mater., 363-365, (2007) 944

Asakura et al, Private communication, 1st Workshop on the “Dust in Fusion Plasmas” (DFP/EPS 2007) Satellite Meeting of the 34th EPS Conference on Plasma Physics Warsaw, Poland (2007).

G.F. Matthews, J. Nucl. Mater., 337-339, (2005), 1.

P. Dore and E. Gauthier , J. Nucl. Mater., 363-365, (2007) 1414.

C. Voinier, C.H. Skinner and A.L. Roquemore, J. Nucl. Mater., 346 (2005), 266.

J. Worms, F. Onofri, C. Grisolia and S. Rosanvallon, Private communication, Workshop on the “Dust in Fusion Plasmas” Satellite Meeting, 34th EPS Conference on Plasma Physics, Warsaw, (2007)

N. Bekris, J.P. Coad, R.-D. Penzhorn, S. Knipe, L. Doerr, R. Rolli and W. Nägele, J. Nucl. Mater., 337-339, (2005), 659-663.

Ph. Delaporte, A. Vatry, D. Grojo, T. Itina, K. Gouriet, M. Sentis, C. Grisolia, S. Rosanvallon, Private communication, Workshop on the “Dust in Fusion Plasmas” (DFP/EPS 2007) Satellite Meeting, 34th EPS Conference on Plasma Physics, Warsaw (2007)

C H Skinner and G Federici, *Phys. Scr.* T124 (May 2006) 18-22.

S. Rosanvallon, C. Grisolia, P. Delaporte, J. Worms, F. Onofri, S.H. Hong, G. Counsell, J. Winter, to be published in J. Nucl. Mater., presented at the ICFRM conf. in Nice, 2007.

B. Patel and E. Letellier, EFDA Report (task JW3-FT-5.12) “Experimental Determination of parameters relevant to radiological safety of JET tritiated dusts”.

7 LESSONS LEARNT ON TRITIUM AND WORKERS IN FUSION DEVICES

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ITER, France

7.1 Introduction

Once the origin of the energy produced by the Sun was understood, the way to reproduce fusion on Earth has always seemed simple. In fact, the problem of fusion has never been the reaction itself, easy to obtain if the energy that is going to move the hydrogen nuclei closer is large enough. The actual problem for years has been the confinement and control of the plasma formed by the hydrogen isotopes before fusion. In the Sun the gravitational field confines and controls the mass of elements, which fuse; on Earth, confinement has been obtained in magnetic traps. The perfection of the magnetic trap has been the fundamental goal of plasma physics and fusion technology research during the last 40 years of the twentieth century. Large scientific and technological advances have helped to achieve this goal. The result is an international project, which gathers the efforts of the entire world research centres involved in this field: ITER.
Lessons learnt in fusion science come from a lot of machines in the world (see table 1), and are also applied to new fusion experimental machines that will not use tritium (see table 2) nevertheless only two installations based on magnetic confinement have used up to now deuterium and tritium and have eventually demonstrated that the fusion reaction are achievable in such machines: the **T**okamak **F**usion **T**est **R**eactor, TFTR in Princeton, USA and JET, the European tokamak.
This article summarises the feedback on Tritium management with regards to the workers in Fusion installations (chapter 2); then the application to ITER design and disposals for its future operation is explained through radioprotection approach, retained means for Tritium confinement and Safety Objectives for ITER (chapter 3). In the conclusion possible improvement for gathering ITER relevant experience in other installations using tritium are suggested.

Table 1: Mains tokamaks contributing to ITER design

Tokamaks	Country	Start	Shut-down	I_p (kA)	R_o (m)	r (m)	B_T (T)	Main Observations
TM-3	USSR	1973	+	30-70	0.4	0.08	1.0 - 3.5	First Te measurement Disruption anomalous transport
TFR	France	1973	1986	150	0.98	0.25	3.0 - 4.0	τ_E measurement. Runaway beams
TUMAN-3	Russia	1973	X	150	0.53	0.22	0.7-0.8	Ohmic H-mode
T-10	Russia	1975	X	450	1.5	0.38	4.	H-mode. Internal transport barriers.
D-III-D	USA	1977	X	2000	1.7	0.7	2.1	H-mode Internal transport barriers. Advanced tokamaks scenarios
T de V	Canada	1980	1998	250	0.83	0.22	2	H-mode
ASDEX	Germany	1980	upgraded	400	1.65	0.4	2.8	X-point/H-mode
ALCATOR C-MOD	USA	1980	X	1000	0.67	0.22	5-8	High field compact experiment. H-mode
TFTR	USA	1982	1997	3000	2.52	0.87	6	Neoclassical tearing modes bootstrap current **DT experiments** Reversed field shear H-mode
JET	EU	1983	X	3000-7000	2.96	1	2.8-4	H-mode Reference Scenarios Advanced tokamaks scenarios **DT experiments** /Remote handling
JT-60 / JT-60U	Japan	1985	X	6000	3.3	1	2.32 4.72	X-point. H mode Advanced tokamaks scenarios
T-15	USSR	1988	X	1800	2.45	0.7	3.6	Superconducting coils
TORE-SUPRA	France	1988	X	700-1300	2.25	0.70	2.09-3.95	Superconducting coils Long pulses technology H-mode
ASDEX-Upgrade	Germany	1991	X	1600	1.65	0.5	3.1	First H-mode. Divertor. ITER-like first wall
T-7/ HT-7	China	1994	X	210	1.22	0.28	2	Superconducting coils-

Table 2: Parameters of new tokamaks in the world and ITER

SST-1	India	2002			1.1	0.2	3.	Superconducting coils
EAST-(HT-7U)	China	2003		1000	1.7	0.4	3.5	Superconducting coils - Steady State plasma
KSTAR	Korea	2003		2000	1.8	0.5	3.5	Superconducting coils X-point
ITER	International	2016		15000	6.2	2	5.3	Superconducting tokamak experimental reactor **DT experiments**
X-operating device, + closed device								

7.2 Tritium management and workers in existing fusion devices

ITER will be operated by the ITER Organization (IO) and it will be licensed in France following European and French regulation. ITER will follow the first process for licensing a fusion facility in the framework of an experimental device with a total Tritium inventory on site of about 3 kg. The main ITER parameters are far from those expected in the future demonstration reactors where the fusion power will be at least 5 times higher than in ITER and the additional heating power could also reach up to 5 times the one foreseen in ITER. Actually ITER is the intermediate stage between the existing machines and those future reactors, DEMO, to which it will provide input on plasma performances, tritium breeding, blanket/divertor designs and solution of engineering issues, as well as bounding accidents or classification of waste and general safety approach [1].

In Magnetic Confined Experiments (MCE), although tokamak fusion research has concentrated upon resolving the issues for power production, most experiments have not used tritium, due to the requirements of handling the radioactive gas and of dealing with the 14 MeV neutron activation [2].
Tritium laboratories for fusion research also play an important role in understanding exposure of the workers to tritium, nevertheless the combination of external exposure due to material activation by fusion neutrons and internal exposure due to tritium inhalation is only possible in fusion devices.
Only two fusion machines in the world have been prepared for tritium handling and have been involved in safety and licensing procedures: TFTR and JET.

In 1991 JET performance had reached the level which warranted the use of tritium for the first time in a laboratory plasma experiment. A peak fusion power of 1.7 MW was produced, averaging 1 MW over a 2 second period. This was the world first controlled production of significant fusion power. In 1993, TFTR became the first tokamak experiment to use a 50:50 deuterium-tritium mixture resulting in 11.5 MW of fusion power [3]. In 1997, 16 MW of fusion power was obtained in JET, and alpha particle heating clearly observed [4]. Since then T-traces have been used in JET for tritium transport plasma studies [5].

7.2.1 Tritium management and workers

Although ITER is the next step in fusion progress the challenge of its expected performances is easy to understand if the pulses in ITER are compared to those in JET and TFTR.
Figure 1 represents a well know comparison of DT pulses in JET and TFTR.
In this figure main fusion parameters are represented: the abscise gives the duration of a pulse in seconds (s) and the ordinate the fusion power during the pulse in megawatts (MW).
On each curve the fusion power gain Q is given

$$Q = P_{fus}/P_{ext}$$

Where
P_{fus}= fusion power as the result deuterium-tritium (D-T) reactions
$[({}^{2}_{1}H^{+} + {}^{3}_{1}H^{+} \rightarrow {}^{4}_{2}He^{2+}$ (3.6 MeV) + n (14.1 MeV)]
P_{ext}=External power for plasma generation, heating, fusion triggering and sustaining
When $P_{ext} = 0$ or $Q = \infty$ then fusion " ignition" is reached.
For reactor conditions Q should be at least 30-40 and alpha particles from D-T reactions are participating to the plasma heating in 85-90%. This condition is called a burning plasma.
When Q~1, output power equals input power and this condition is called "scientific break-even".

In figure 1 it can be observed that JET transiently approached Q~1 in a pulse of a couple of second reaching 16 MW.

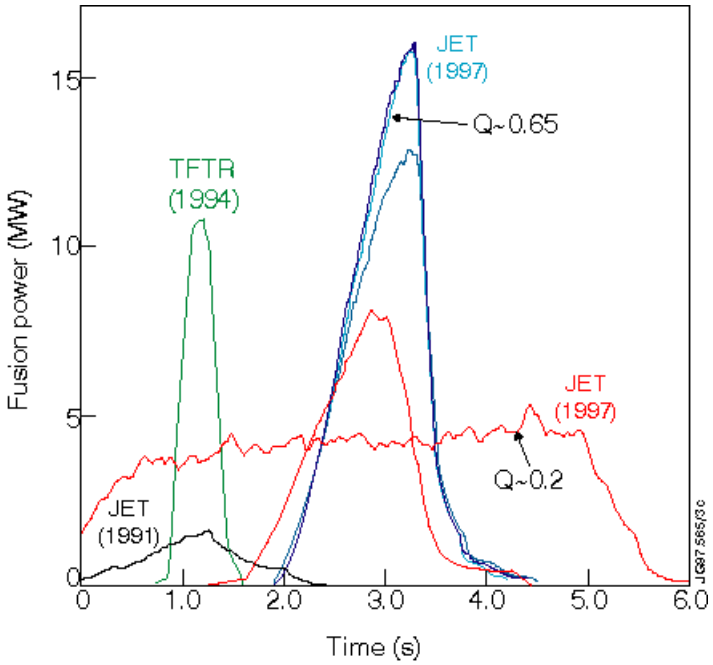


Figure 1: Deuterium-tritium experiments in TFTR and JET.

For longer pulse (~6s) Q was much lower (0.2) for a power of 4 MW. Compared with this "long pulse", ITER will multiple by 100 the power en the length of its pulse and by 20 the fusion power Q, achieving a so-called an "extended burn", Q=10 and heating by alpha particles of the order of 60%. (see figure 2). Obviously a big difference will also be the amount of tritium on site.

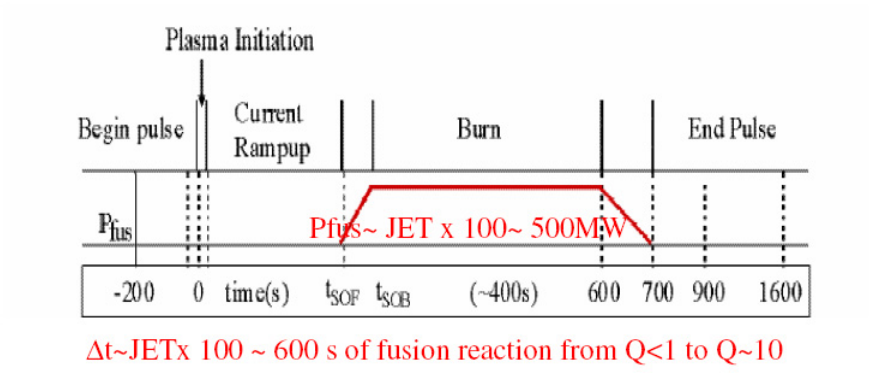


Figure 2: Fusion power evolution in an ITER pulse. Compared to JET Power and time duration are multiplied by 100.

7.2.1.1 DT JET experiment

A safety case for JET D-T operation was required before starting DT experiments. UK requirements and standards were followed. This corresponded to a formal approval for tritium operation for the Active Gas Handling System (AGHS) and for the torus systems. [6]

Safety principles were identified. Accident sequences were analysed both with deterministic and probabilistic methods. Results in terms of source terms, on-site and off-site doses were carried out. Although an extension for JET has been recently approved in the scope of the Broad Approach and JET ITER-relevant experiments the dismantling of JET in part of the UKAEA decommissioning safety [7]. The following amount of tritium (table 3) on site have been maintained from traces from commissioning phase to higher values in DTE experiments in a progressive evolution of the experiment and inventories that will be followed also in ITER.

Table 3: Tritium on site at JET

Operation phase	**year**	**tritium on site**
Hydrogen	1984	0
Deuterium-Deuterium	1986	0
Deuterium-Deuterium plus Beryllium	1989	0
Deuterium-Tritium (PTE)	1991	0.25g
Divertor shutdown	1992	0.1g
AGHS trace tritium commissioning	1995	0.1g
AGHS full tritium commissioning	1996	3g
Deuterium-Tritium (DTE1)	1997	20g
Trace tritium experiment	2003	<10g

Concerning the exposure of the workers to ionising radiation, studies have been done since the first experiments involving tritium [for example 8, 9, 10]. JET policy has been to sample all persons making access to tritium handling areas. Measured committed effective doses have been described by intervention teams giving collective doses and by individual doses. It must be underline that collective doses before deuterium-tritium campaign have been by 1988 of the same order that after deuterium-tritium campaign before1997, still staying very low. The main contribution to the doses has always been activations of structural material of the tokamak. Before 1997 the long-term average collective dose was 96 p-mSv/a and average individual worker dose ~0.150 mSv/a. After 1997 the average collective annual dose has been 30 p-mSv/a and average individual dose 0.058 mSv/a [10]. Since 1997, the contribution of the internal doses due to Tritium inhalation to the total doses has been of the order of 2%. A decreasing in the total doses of the order of 50% has been clearly related to the use of remote handled tools for maintenance since 1998, a decrease of a about 50% was also observed for the internal doses. The following points may be summarised for JET:

- *Since 1998 Remote handling has been used for maintenance drastically reducing total doses to the workers*
- *The tritium dose has been of the order of 2 % of the total worker dose*
- *The group responsible for all machine maintenance and repair work has been the most exposed group*
- *About 75% of collective dose was due to maintenance*
- *1/3 of collective dose have been measured on non-maintenance workers*

The success of reduction of doses to the workers is clearly related to implementation of radiation protection measures for reducing the total doses such as

- *ALARA in operation*
- *Remote handling*
- *Radiological zoning*
- *Ventilation and detritiation systems in AGHS*
- *Protection suits*

In chapter 3 the extension to ITER will be detailed.

7.2.1.2 DT TFTR experiment

TFTR safety analysis process consisted of a Preliminary Safety Analysis Report (PSAR) developed in 1976-1977, and approved by the US Department of Energy (USDOE) in 1978. In the US method, PSAR approval is needed before any site construction work could proceed. The Final Safety Analysis Report was written in 1980-1982, and approved by the USDOE in 1982. An FSAR amendment for deuterium-tritium operations at TFTR was presented to the USDOE and approved in 1992.

During the DT campaign in TFTR, the tritium fuelled the plasma either by gas puffing or by tritium neutral beam injection. For than 3 years, about 37 PBq of tritium (100 g) had been processed while maintaining a 5g site limit. During the last 3 months of TFTR operation, the tritium was processed on site, with a tritium purification system. During this time and also during the post operation shutdown, the radiation doses to PPPL workers were maintained at pre-tritium levels [11]. The key factors, which allowed this safety record, were thorough documentation of the installed hardware and careful planning of all activities. The TFTR Decontamination and Decommissioning (D&D) Project started at the beginning of October 1999. The last commitment of the TFTR Project was the removal and safe disposal of the TFTR device. The TFTR D&D Project was completed in three years in time and cost [12].
Data in [11] on individual and collective doses at TFTR show a higher contribution of tritium to the total doses, in average 9%, although only on-fifth of the exposed workers were monitored. No remote handling was available on TFTR.

7.3 Application of lessons learnt to ITER radioprotection

ITER will be licensed in France as a Basic Nuclear Installation (Installation Nucléaire de Base, Laboratoires et Usines). Consequently French Nuclear regulation applies to all the aspects of design, construction, operation and dismantling. It applies in particular to the protection of the workers against ionizing radiations which is considered as a safety function.
Besides design principles following IAEA Safety Series No. 110 on safety for nuclear installation have been applied: The exposure to radiation of site personnel and releases of radioactive materials to the environment shall be made by design As Low As Reasonably Achievable (ALARA) and the systematic consideration of the man–machine interface and human factors shall be included in all stages of design and in the associated development of operational requirements.
The means defined in design phase aim to protect against either external or internal exposure and are separated into collective and individual protective measures. ITER has set objectives for individual and collective doses below statutory doses limit and will stay below the objectives as indicated in table 4.

Table 4: General Safety Objectives for ITER

Condition	Personnel		Public and Environment	
	General Safety Objective*	Consequence Analysis	General Safety Objective*	Consequence Analysis
Design Basis Situations				
Normal (per year)	As Low As Reasonably Achievable, and in any case less than: < 10 mSv/a	< 5 mSv/a	ALARA and in any case ≤ 0.1 mSv/a	< .01 mSv/a
Incident (per event)	As Low As Reasonably Achievable, and in any case less than < 10 mSv	Managed per event	< 0.1 mSv per incident.	<< .01 mSv per event
Accident (per event)	Consider occupational exposure management of accidents.	Managed per event	< 10mSv No restrictions on the consumption of produce or meat.	< 2 mSv Early dose at site boundary < 1 mSv Long-term dose at 2.5 km
Beyond Design Basis				
Hypothetical Accidents	Consider occupational exposure in management hypothetical situations.		No cliff-edge effects; possible countermeasures limited in time and space.	< 10 mSv

General Safety Objectives were presented to French Safety Authorities in Dossier d'Options de Sûreté (DOS) and reviewed by Safety Authorities November 2002.
These objectives are the same in Rapport Préliminaire de Sûreté (RPrS) which will be submitted to the French Safety Authorities at the end of 2007.
The individual dose objectives set for ITER operations are a maximum individual dose below or equal to 10 mSv/yr, the mean individual dose being below of equal to 2.5 mSv/yr.

Neither domestic nor European regulations stipulate a collective dose limits. Various data can nevertheless be referred as a feedback for ITER collective dose:

- the maximum annual collective dose objective set by EDF in 2002 for each power reactor unit was 1000 person mSv/yr,
- the new objectives set by the US Institute of Nuclear Power Operations in 2005 were 650 person.mSv/yr for pressurised water reactors (PWRs) and 1200 person mSv/yr for boiling water reactors (BWRs). In France, the 2005 objective was 800 person.mSv/yr for pressurised water reactors.
- The collective dose objectives set for CANDU reactor unit shutdown periods (heavy water reactors that produce large quantities of tritiated water) are of the order of 750 to 1100 person mSV for maintenance operations.

Then the maximum annual collective dose objective, taken as an average throughout the ITER operating lifetime, is set at 500 person mSv/yr.

7.3.1 ALARA

In addition to statutory dose limits ITER approach for worker protection is based on a constant revision of design and operation to ensure that hazards are maintained As Low As Reasonable Achievable (ALARA). ALARA approach for ionizing radiation is extended to chemical exposure risk to Beryllium in the design and in operation.
For ITER design different ways for optimization have been used and ALARA will be applied also in operation.

7.3.2 Ventilation and detritiation systems in AGHS

From lessons learnt in previous fusion and tritium laboratories installations, ITER has improved design for tritium confinement. Main worker protection against tritium internal exposure and against the risk of spread of radioactive substances is assured by a coherent set of physical barriers and auxiliary techniques intended to confine radioactive substances. As shown in figure 3, these confinement systems can be static: a vessel, a wall, a glove box and can include more than one physical barrier against releases; a dynamic system, ensures a preferential airflow from the least contaminated zones into the most contaminated zones and provides at the same time an additional air decontamination and renewal function by a detritiation network. The primary confinement system is designed to prevent releases of radioactive materials into the accessible working areas, the secondary confinement system prevents releases of the contamination to the working areas accessible by non-authorized radiological workers, the general public and the environment. Primary and secondary systems include detritiation means where a risk of tritium spread is not excluded. This approach for secondary systems is a new approach which will be implemented in ITER supported by previous experiences in JET and TLK and in R&D for new technologies.

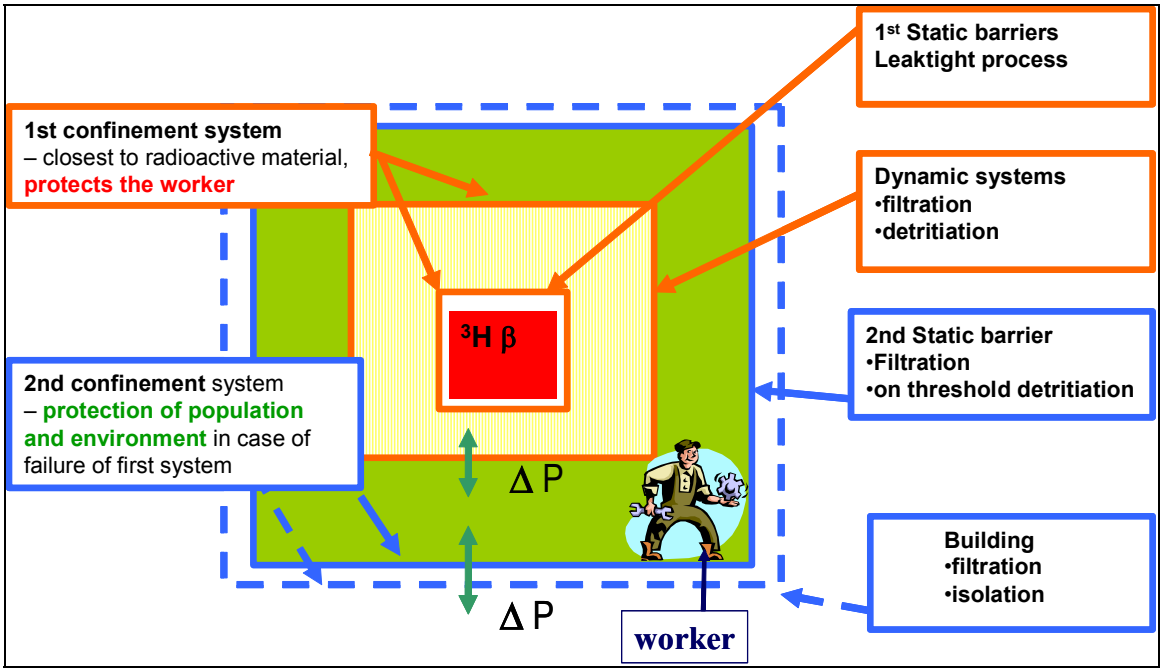


Figure 3: Example of static and dynamic confinement systems

7.3.3 Remote handling

As shown at JET, in ITER the main contribution to the doses to the workers has always been activations of structural material of the tokamak. In order to minimise these doses, ITER will extensively use remotely handled tools. In figure 4 an artistic view of part of ITER tokamak complex which includes tokamak building, tritium building and diagnostic buildings is shown.

In the centre the tokamak device is designed surrounded by a concrete bio-shield of 2 meters thick. During the assembly phase of the tokamak all the internal components will be mounted using remotely controlled robots based in the experience at JET in particular of full mounting and dismounting of part of the machine (divertor). The calculation of activation for dose rates have taken at the end of life of ITER in order to consider the more stringent workers conditions leading to the radioprotection zoning.

- Material choice for reducing contribution to the dose to operators
- Activation for exposure conditions
- Maintenance activities in locations around the tokamak wait 11.5 days after shutdown (10E+6 seconds)➔Dose rate behind bio shield <10 μSv/h
- Activities in the Tokamak Cooling Water System (TCWS) vault wait 5 days after shutdown (<10 μSv/h)
- Activities in port plug and VV by Remote handling
- Optimizing of human intervention in other areas
- Controlled access and circulation of personnel and radioactive materials
  - No access to Tokamak building during operation
  - No access during remotely operated cask transfers.

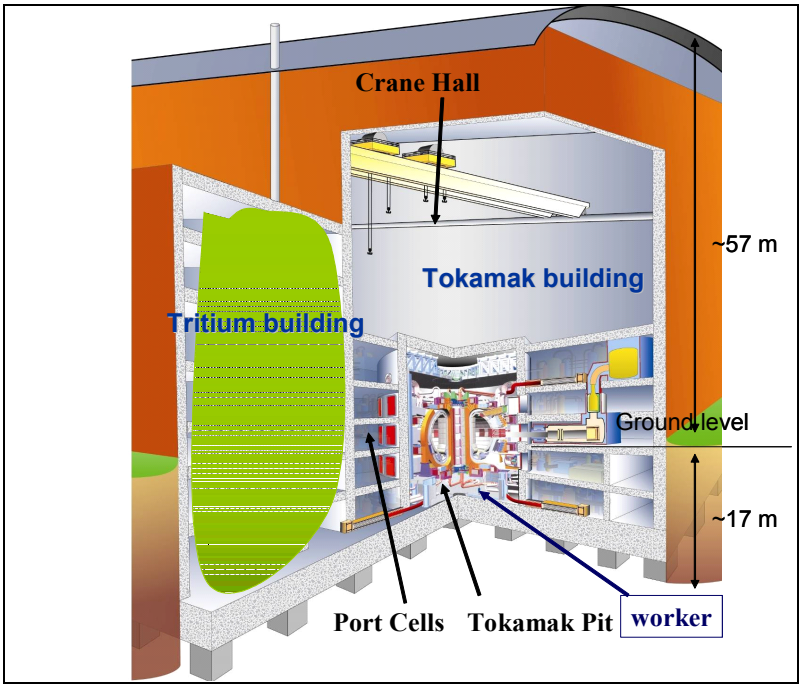
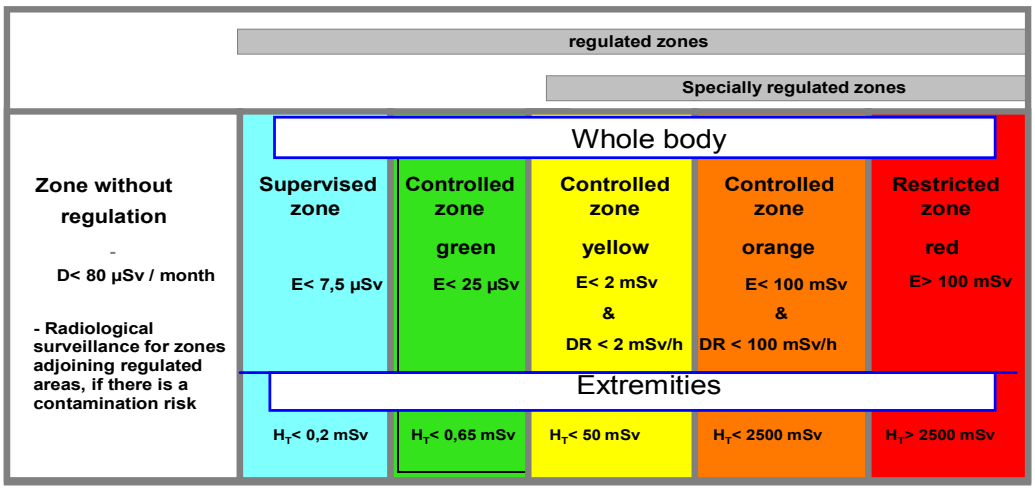


Figure 4: Artistic view of part of ITER tokamak complex which includes tokamak building, tritium building and diagnostic buildings.

7.3.4 Radiological zoning

For radiological zoning, the activation of material at the end of life of ITER has been considered although the neutron fluence will increase along the 20 years of experimentation. Internal contamination has also been considered. In application of French regulation the Order of 15 may 2006 has been used for defining ITER radiological zoning, which is summarized in the table below where E is the potential effective dose in one hour, DR the dose rate, and H the equivalent dose in one hour:



For air contamination there is no reference values in the decree and internal and external doses are equivalence. It is indicated that zoning should be determined considering no use of individual protection devices and an operational derived value is proposed as the average equivalent dose rate on the duration of the operation < 25 µSv/h. In ITER the operational derived value has been defined taking into account the skin transfer as
1 ODV = 25E-6 [Sv/h] / (1.2 [m3.h] * 2.7E-11 [Sv/Bq])= 7.7E5 [Bq.m-3] .
An example of the radiological zoning for tokamak building is given in figure 5.

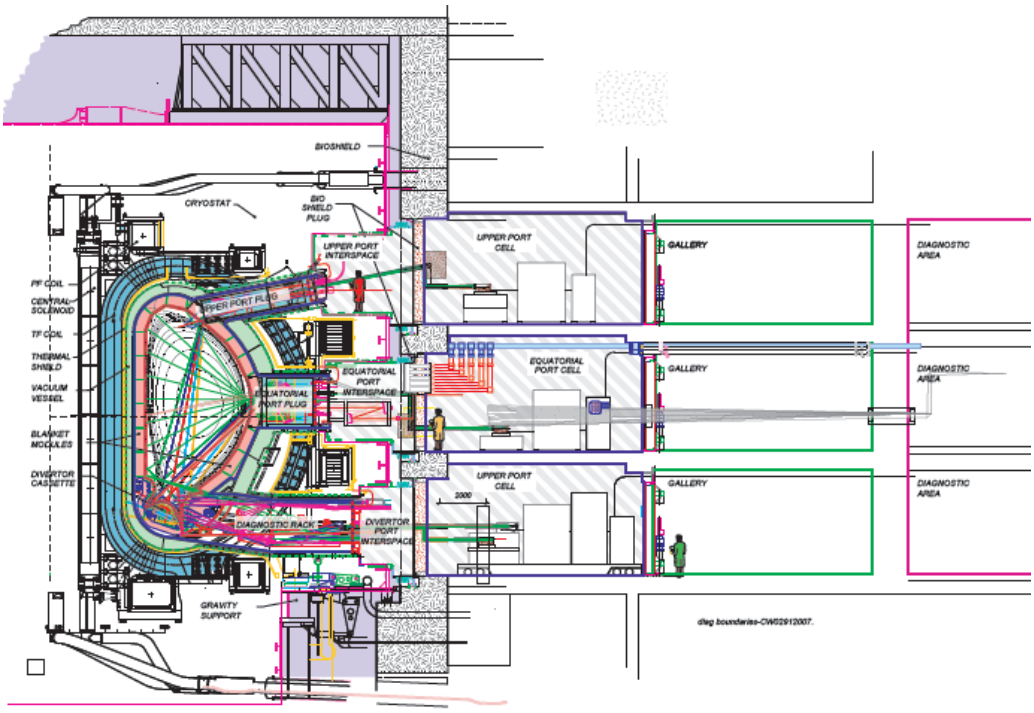


Figure 5: Radiological zoning of the tokamak building: Galleries are green zones, equatorial port cell yellow zones. Access is not allowed inside the bio shield.

7.4 Conclusions

ITER, which will demonstrate the scientific and technical feasibility of magnetic confinement fusion, represents a large step forward from the science level to the energy development one, since it fulfils both fusion plasma conditions and energy development conditions.

- The experience on Tritium and workers in fusion devices is available
  - Tritium had an impact of about 1-2% of the collective dose to the workers at JET
  - In fusion devices 20%-25% of the doses to the workers were on non-maintenance teams
- The key factors for low doses ALARA approach radioprotection measures, remote handling, thorough documentation of the installed hardware and careful planning of all activities.
- Technologies for Tritium confinement and detritiation (atmosphere and water) system for Tritium recovery are well developed in for new devices.
- More feedback may be gathered from Tritium laboratories and CANDU reactors
- ITER will be the first fusion machine fully designed for operation with equimolar DT
- European and French regulation will be applied also for radioprotection.

7.5 References

[1] MAISONNIER D. et al., The European Power Plant Conceptual Study, Fusion Engineering and Design.

[2] RODRÍGUEZ-RODRIGO L. et al., Summary report on the operation feedback in the existing fusion machines. EFDA contract report EISS-GAC2-DEL-2001-00041.

[3] STRACHAN, J. D. et al. "TFTR DT experiments". Plasma Phys. Control. Fusion 39, B103 (1997).

[4] KEILHACKER et al., " High fusion performance from deuterium-tritium plasmas in JET " Nucl. Fus. **39**, 209, (1999).

[5] ZASTROW K-D, Tritium transport experiments on the JET tokamak, Plasma Phys. Control. Fusion 46 (2004) B255–B265.

[6] BELL A.C., The safety case for JET DT operation. Fusion Engineering and Design, **47** (1999).

[7] UKAEA http://www.ukaea.org.uk/downloads/corporate/ukaea_ds/Vol_2_Part4.pdf.

[8] PATEL B. et al. Health physics aspects of tritium operation at JET. Fusion Engineering and Design **47**, 267 (1999).

[9] BELL A. C. et al. Tritium inventory control the experience with DT tokamaks and its relevance for future machines. Fusion Engineering and Design **66-68**, 91 (2003).

[10] NATALIZIO A. et al. Collection and analysis of occupational radiation exposure data from the JET tokamak. Fusion Engineering and Design **75-79**, 1193 (2005)

[11] NATALIZIO A. et al. TFTR occupational radiation exposure data collection and analysis. Fusion Engineering and Design **75-79**, 1187 (2005).

[12]-TFTR decommissioning completed on schedule, Fusion Power Report-02-07 October 2002.

8 SUMMARY

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In consultation with the Working Party "Research Implications on Health and Safety Standards" of the Article 31 Group of Experts*

8.1 Introduction

This document summarizes the presentations and, through the conclusions, tries to emphasize the potential implications of the Scientific Seminar on Emerging Issues on Tritium and Low Energy Beta Emitters, held in Luxembourg on 13 November 2007. While it is not intended to report in an exhaustive manner all of the opinions that were expressed by the speakers or by the audience, it takes account of the discussions that took place during the seminar and the subsequent meeting of the Article 31 Group of Experts on 14 November 2007. The content of the document has been submitted to the lecturers and discussed within the Working Party RIHSS (Research Implications on Health Safety Standards).

8.2 Rationale of the Scientific Seminars

The European Commission organises every year, in cooperation with the Group of Experts referred to in Article 31 of the Euratom Treaty, a Scientific Seminar on emerging issues in Radiation Protection – generally addressing new research findings with potential policy or regulatory implications. Leading scientists are invited to present the status of scientific knowledge on the selected topic. Based on the outcome of the Scientific Seminar, the Group of Experts referred to in Article 31 of the Euratom Treaty may recommend research, regulatory or legislative initiatives. The European Commission takes into account the conclusions of the Experts when setting up its radiation protection programme. The Experts' conclusions are valuable input to the process of reviewing and potentially revising European radiation protection legislation.

8.3 Background and Purpose of the 2007 Seminar

In 2007, the Scientific Seminar addressed "Emerging issues on tritium and low energy beta emitters". Experts reported on the relevance of dose for low energy beta emitters, on metabolism, radiobiology and epidemiology of tritium, on tritium in the environment (sources, measurements and transfer), and on tritium in fusion facilities. The seminar raised a number of issues that merit further attention such as the biological impact of incorporated tritium, which may have to be reconsidered given the new data on risk from organically bound tritium. The seminar pointed to the need for further research, for example in epidemiological studies on the effects of tritium, biotransformation and food accumulation (particularly of organically bound tritium) effects in early pregnancy, and the impact of tritium particulates.

* The following members of the Working Party Research Implications on Health and Safety Standards of the Article 31 Group of Experts contributed to the preparation of this document: L. Lebaron-Jacobs, W-U Müller, P. Olko, S. Risica, P. Smeesters (Chairman of the WP), R. Wakeford.

Over the past fifteen years recurrent questions arose concerning tritium dosimetry and health hazards:

- Is the tritium beta particle more deleterious than X-rays or ^{60}Co gamma-rays?
- What proportion of tritium becomes OBT after incorporation in the organism?
- What are the consequences of tritium contamination on the embryo?

Recent experimental and epidemiological data do not materially challenge the assumption of the comparatively low level of tritium radiotoxicity. The experimental results agree on a hazard regarding acute OBT contamination of about two or three higher than that with HTO: it is included in the dose coefficients. Data on chronic OBT or HTO contamination are very few and do not allow to provide a specific quantitative risk assessment. Some recent analyses suggest a hazard two-fold higher than for an acute exposure. Finally, discussion about the RBE took place: 1 or 2?
Several important factors must be considered when looking at the effects of age on anatomical, physiological and biokinetic data. Also, there is no current dosimetric model for calculating doses to the embryo/fetus resulting from placental transfer of radionuclides following intakes by the mother. Without additional knowledge of radionuclide transport across the placenta and the associated distribution in the embryo/fetus at different times during development, it will be difficult to set and justify regulatory limits for tritium exposure for pregnant women.
The functioning of fusion reactors requires tritium and generates tritium. In the future reactor ITER, exposure of workers is possible, and more precisely as tritium metallic dusts, whose biological behaviour is unknown.
Four main issues emerge from the analysis of tritium dosimetry and health hazards:

- the relation between the low-energy beta-emitters and the RBE
- the characterization of the risks from chronic exposures to OBT and HTO
- the characterization of hazards in conditions of maximum radiosensitivity (the embryo/fetus) after an acute or a chronic exposure to HTO or OBT
- the biological behaviour, the biokinetics, the effects of tritiated dusts or particles relative to other toxic exposures (occupational exposures in both the fusion and fission fuel cycles).

The purpose of 2007 Seminar was to highlight the questions raised by the exposure to tritium (tritium metabolism, dosimetry and biological effects, environment) and to know more about the potential exposure of workers in future fusion devices.

8.4 Main Points Arising from the Presentations

Dudley T. Goodhead - *The relevance of dose for low-energy beta emitters*

In his presentation Prof Dudley Goodhead emphasized on the specific features of tritium such as:

- increased average ionization density on subcellular scale (track-average LET (Linear Energy Transfer) of 4.7 keV/μm for tritium and of 0.22 keV/μm for ^{60}Co)
- short range of electrons (beta-particles) leading to non-uniformity of dose when emitters are inhomogeneously distributed (OBT)
- non-uniformity of absorbed dose (tritiated DNA precursors, OBT in adipose tissue)
- larger mean energy deposition by single tritium beta than from single track from ^{60}Co gamma (fewer cells or nuclei are hit by tritium, but they are hit harder)
- nuclear transmutation (conversion of ^{3}H to ^{3}He loses its chemical binding in molecule)
- isotopic mass difference ratio compared to stable isotope affects physico-chemical properties (differential diffusion)

- different molecular forms (tritiated water (HTO), organically bound tritium (OBT)).

D. Goodhead specified that most of these features are not incorporated into conventional radiation protection dosimetry, but they can be integrated in various ways into experimental measurements of RBE. He added that it may be expected that tritium beta-decay will produce a greater yield of double strand breaks (DSB) than the same absorbed dose of ^{60}Co gamma rays and a higher proportion of complex DSB, both of which factors may lead to greater biological effectiveness. The problem is to establish the likely effectiveness of low-energy beta emitters for human risk relative to reference radiation: most of the literature, even to recent times (e.g. ICRP 2003) refers to small experimental data from Bond et al. (1978), which are of questionable relevance to human risk (data on dicentric chromosome aberrations in human lymphocytes irradiated in vitro).
He recommended:

- using available information to establish the likely effectiveness of low-energy beta emitters for human risk relative to reference radiation
- considering special cases of potential practical relevance (e.g. extreme inhomogeneity)
- experimental determination of yields and complexities of DNA damage from tritium and beta emitters in comparison with reference radiation
- seeking agreement on single reference radiation of practical convenience and relevance to established human risk
- applying a raised RBE of 2 to all low-energy internal emitters (β emitters, soft X-ray emissions and Auger emitters) excluding external irradiations with soft X (very high attenuation as they pass into the body).

John Harrison - *Doses and risks from tritiated water (HTO) and environmental organically-bound tritium (OBT)*

John Harrison, from the Health Protection Agency (HPA, UK), discussed current assumptions and uncertainties about the dose coefficients for HTO and OBT. The models provided by the International Commission in Radiological Protection (ICRP) consider intakes of HTO and OBT by ingestion and inhalation by adults and children and doses to the fetus following intakes by the mother. However, the models make a number of simplifying assumptions (no dose coefficients for specific forms of OBT (eg. DNA precursors)). Taylor (2003) proposed a new biokinetic model for HTO with three components of retention (half-times of 10 days, 40 days and 350 days) that will be used in future ICRP calculations.
Greater uncertainties in dose coefficients for OBT arise from uncertainties from the fraction incorporated into OBT in body tissues and the associated retention times since they are based on animal data. ICRP dose coefficients are provided as point values without uncertainties; however, the data on which they are based are subject to uncertainties. Hodgson et al. (2005) showed that the ICRP generic dose coefficient for OBT might underestimate dose to fish consumers (50% greater than the ICRP value).
J. Harrison reported that Straume and Carsten (1993) provided a 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. For all observed effects of HTO exposure, relative biological effectiveness (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. This study was confirmed by the data of a recent review by Little and Lambert (2007) obtaining a RBE estimate of 2.19 (95% CI 2.04, 2.33) with respect to chronic gamma radiation and 1.17 (95% CI 0.96, 1.39) with respect to chronic X-irradiation. However, RBE data were obtained using radiation delivered at higher doses and dose rates than those generally received by workers or the public. Consistent with these reviews, an Advisory Group to the UK HPA (AGIR) has recommended:

- the use of a standard high energy □-ray source such as 60Co as a reference for RBE studies

- an RBE value of 2 for tritium epidemiological studies and retrospective dose assessments
- ICRP to consider adopting a value of 2 for the tritium radiation weighting factor (wR).

J. Harrison has drawn attention to the relationship between DDREF and RBE. A DDREF of 2 is applied by ICRP in considering risks of solid cancers in humans (Japanese survivors). The greatest RBE values for tritium will be seen for experimental end-points for which the test radiation exhibits the greatest DDREF. Since the seminar, Cox et al (2008) have clarified the ICRP position that a w_R of 1 will continue to be used in the calculation of the ICRP protection quantities. ICRP are preparing further advice on the application of equivalent and effective dose, including assumptions that might be made when the protection quantities are not applicable and best estimates of dose and risk are required.

Barrie Lambert - *Radiobiology and epidemiology associated with exposure to tritium*

Barrie Lambert is a member of the AGIR subgroup on Tritium Risks. He presented both the radiobiological issues and the epidemiological ones. He specified the reproductive effects of tritium in the female and discussed the results on RBE studies.
During pregnancy tritium will be incorporated into most female tissues and also into fetal oocyte DNA and other organic fractions during the five-month period from two months after fertilization until seven months of gestation. Tritium could in principle remain in fetal oocytes until fertilization decades later. He reported the results of a study on a critical group consuming Cardiff Bay flounder during pregnancy: approximately 4% of oocytes could experience a tritium disintegration within 30 years. The frequency of severe hereditary effects resulting from this would not be significant, around 10^{-6} compared to spontaneous incidence, around 3 to 4 %.
B. Lambert explains that in the literature a range of reference radiation have been used for the determination of RBE values of tritium. However differences have been observed in the biological effectiveness with reference to the X-rays and gamma rays: the values obtained when the reference radiation was ^{60}Co gamma rays were greater than those obtained when the reference radiation was orthovoltage X-rays. Moreover, the interpretation of RBE experiments is complicated by the fact that dose rates are rarely comparable and the reference radiation may itself be more effective than hard gamma rays. Finally published RBEs could underestimate the actual RBEs relevant for human risk assessment since many of the studies employed radiation delivered at higher doses and dose rates than those generally received by people.
B. Lambert provided a review of epidemiological studies. In his opinion, the most robust analyses were the nuclear worker studies in Canada, Savannah River Site and possibly other sites in the USA, and finally the five main tritium-exposed UK nuclear workforces (Sellafield, Chapelcross, Capenhurst, at the AWE and the UKAEA): the dosimetry was apparently good.

B. Lambert concluded that:

- the frequency of severe hereditary effects, resulting from the analysis of the critical group consuming Cardiff Bay flounder during pregnancy, is non significant compared with spontaneous incidence,
- various theoretical and experimental studies with radiation of LET similar to that tritium beta particles has led to the general expectation of an RBE of at least 2 for tritium compared with gamma radiation,
- in general, the available epidemiological studies do not contain enough detail to estimate risks from tritium.

Philippe Guétat - *Tritium and the environment: sources, measurement and transfer*

Philippe Guétat is from CEA, Valduc (France). He presented the different sources of tritium, different methods of measurement associated to tritium processes and survey, transfer to the biosphere compartments in normal and accidental conditions.
He specified that natural tritium production is higher than man-made release. Main sources of releases come from reprocessing plants and heavy water reactors. He indicated that waste inventory is very small but inappropriate limit for tritium in water table generates a need of interim storage. He pointed out that fusion technology will increase the use of tritium but should not change fundamentally release levels. However, treatment conditioning of waste may be a significant source. In case of accidental release of tritium, tritium does not concentrate in food chain, but can remain in soil during an agricultural season. He concluded that experiments are needed because of uncertainties about wet and dry deposition velocity onto soils, the uptake by plants of HTO from air and soil during the night, and the translocation of OBT throughout the plant.
P. Guétat concluded that:

- tritium has to be considered apart from the other beta-gamma elements because of its low radiotoxicity and its difficult confinement
- the concentrations in foodstuffs specified by the Codex Alimentarius as intervention levels are too low for tritium
- 100 Bq l^{-1} should be kept as an investigation level, but not misunderstood as the drinkable limit.

Christian Grisolia - *Plasma wall interaction in fusion machine (micro-particle creation processes)*

Christian Grisolia is from CEA, Cadarache (France). He presented the plasma wall interactions in Tokamaks and Tokamak tritium cycle, dust creation processes and ITER predictions, in vessel micro-particle measurements and removal techniques.
The fusion reaction commonly used in fusion reactors is a mixture of Deuterium and Tritium heated to more than 200 Billion degrees. Fusion is observed and helium nucleus is created with a high energy neutron: a huge amount of energy is released.
C. Grisolia specified that one consequence of this plasma outflow interaction with the wall is the sputtering of the materials which form the plasma facing components (PFCs). This erosion leads to the re-deposition of material. Part of the fuel of the discharge is trapped in the deposited layers: it is considered that up to 5 g of tritium could be trapped in these layers per ITER shot. Due to internal stresses, these layers are breakable and generate micro-particles or dust. The AMAD (Activity Median Aerodynamic Diameter) of these particles is shown to be equal to 7 μm. He pointed out that the link between the airborne dust measured and the dust which can be really mobilized relies on complex fluid codes that do not yet exist. However, a set of system seems to be available: regarding the values accepted by ITER guidelines, removal tools will be used.

Lina Rodriguez-Rodrigo - *Lessons learnt on Tritium and workers in Fusion devices*

Lina Rodriguez-Rodrigo is from ITER Organization in Cadarache (France). She reported that lessons learnt in fusion science came from machines in the world such as the Tokamak Fusion Test Reactor (TFTR) (Princeton, USA) or JET, the European Tokamak (UK). Safety principles were identified and accident consequences were analyzed both with deterministic and probabilistic methods. She pointed out that since 1998 remote handling has been used for maintenance in JET drastically reducing total doses received by the workers: the tritium dose has been of the order of 2% of the total worker dose. She identified that in TFTR individual and collective doses showed a higher contribution of tritium to the total doses, in average 9%: no remote handling was available in TFTR. Moreover, 20% – 25% of the worker doses were attributed to non-maintenance teams in fusion devices.

L. Rodriguez-Rodrigo concluded that technologies for tritium confinement and detritiation system for tritium recovery are well developed for new devices such as ITER. However, we need more feedback to be gathered from Tritium laboratories and CANDU reactors.

8.5 Discussion

One question during the discussion addressed the risk exerted by tritiated DNA- (^{3}H-thymidine, for example) and histone-precursors (like ^{3}H-arginine). Quite a number of papers on this topic have been published (see References). Many of these experiments have been carried out with pre-implantation embryos. However, the results are in principle applicable to all cell nuclei. The problem originates from the ability of plants and of some micro-organisms to use HTO for the production of tritiated molecules which may be incorporated into macromolecules of human cell nuclei via the food chain. In this case, the amount of activity taken up by the human body primarily counts, not RBE. The reason is simple: due to the highly heterogeneous distribution, ^{3}H-thymidine, for example, is between 1,000 and 5,000 times more effective than HTO when the same activity is applied. The problem is the short range of the ß-particles emitted by tritium with the result that almost all of the energy is deposited in the cell nucleus.
It is urgently required to draw the attention, in particular, of those people advancing fusion technology to this problem. It might turn out that the conversion of HTO/HT in the environment into cell nucleus seeking compounds is negligible, but this should be checked before substantial releases of inorganic tritium compounds occur.

M. H. L. Snow. Abnormal development of pre-implantation mouse embryos grown in vitro with ^{3}H-thymidine. J.Embryol.Exp.Morph. 29:601-615, 1973.

D. Horner and A. McLaren. The effect of low concentrations of [^{3}H]-thymidine on pre- and postimplantation mouse embryos. Biol.Reprod. 11:553-557, 1974.

S. J. Kelly and J. Rossant. The effect of short-term labelling in [^{3}H] thymidine on the viability of mouse blastomeres: alone and in combination with unlabelled blastomeres. J.Embryol.Exp.Morph. 35:95-106, 1976.

C. Streffer, St. Elias, and D. van Beuningen. Comparative effects of tritiated water and thymidine on the preimplanted mouse embryo during cultivation in vitro. Curr.Top.Radiat.Res.Quart. 12:182-193, 1977.

A. Spindle, K. Wu, and R. A. Pedersen. Sensitivity of early mouse embryos to [^{3}H] thymidine. Exp.Cell Res. 142:397-405, 1982.

L. Clerici, M. J. Carroll, M. Merlini, L. Vercellini, and F. Campagnari. The toxicity of tritium: the effects of tritiated amino-acids on preimplanted mouse embryos. Int.J.Radiat.Biol. 45:245-250, 1984.

W.-U. Müller, C. Streffer, and L. Glück. Radiotoxicity of ^{3}H-thymidine and ^{3}H-arginine in preimplantation mouse embryos in vitro. Rad.Protect.Dosim. 16:155-158, 1986.

W.-U. Müller and A. Spindle. Induction of sister chromatid exchange in preimplantation mouse embryos in vitro by ^{3}H-thymidine or ultraviolet light in combination with caffeine. Teratog.Carcinog.Mutagen. 6:107-114, 1986.

W.-U. Müller, C. Streffer, M. Molls, and L. Glück. Radiotoxicities of ^{3}H-thymidine and of ^{3}H-arginine compared in mouse embryos in vitro. Radiat.Res. 110:192-198, 1987.

W.-U. Müller, N. Heckeley, and C. Streffer. Effects of cell cycle specific exposure to ^{3}H-thymidine or ^{3}H-arginine on development and cell proliferation of mouse embryos. Radiat.Environ.Biophys. 35:267-271, 1996.

9 CONCLUSIONS

Working Party "Research Implications on Health and Safety Standards" of the Article 31 Group of Experts[†]

The radio-toxicity of tritium is considered low, indeed among the lowest of several hundred radio-isotopes. Notwithstanding this, tritium remains of interest in radiological protection for several reasons, not least it is both ubiquitous in the environment (from both natural and artificial sources), its particular features as a very low energy beta emitter when it decays, leading to an inhomogeneous energy deposition at a sub cellular scale, and its potential future significance should nuclear fusion become a major source of energy generation in the 21st century.

There is broad consensus that the current provisions within the system of radiation protection for tritium are broadly adequate subject to the following reservations / refinements:

i) there is increasing evidence that the RBE of tritium is greater than one and that a value of two better reflects the available scientific evidence; consideration should be given at national / regional / international levels to an upward revision in the radiation weighting factor for tritium from one to two, with the objective of improving the coherence of the radiation protection system.

ii) the increased radio-toxicity of organically bound tritium (OBT) – compared with tritium in the form of tritiated water – is well recognised and taken account of within the radiological protection system. Some concerns remain, however, over the relevance of the concept of dose in an organ or tissue in those cases where the distribution of doses is very heterogeneous, in particular where biologically more sensitive structures are preferentially exposed – for example where tritium is incorporated within DNA (e.g. thymidine) or histone precursors (e.g., arginine). Further research into the biological effectiveness of tritium incorporated into such forms, especially during various stages of pregnancy, would resolve these concerns or indicate a need for additional protection measures.

iii) the levels specified for tritium in *CODEX Alimentarius levels for radionuclides in foods contaminated following a nuclear or radiological emergency for use in international trade* were derived generically for application to low energy beta emitters as a group. Significantly, different levels could result had they been derived explicitly for tritium. Given the ubiquitousness of tritium and its increasing importance in the context of fusion energy, consideration should be given to tritium being addressed explicitly in any future revision of CODEX Alimentarius – and of *Euratom Council Regulation laying down maximum permitted levels of radioactive contamination of foodstuffs and of feeding stuffs following a nuclear accident or any other case of radiological emergency*.

[†] The following members of the Working Party Research Implications on Health and Safety Standards of the Article 31 Group of Experts contributed to the preparation of this document: L. Lebaron-Jacobs, W-U Müller, P. Olko, S. Risica, P. Smeesters (Chairman of the WP), R. Wakeford.

iv) fusion is expected to make an increasing contribution to energy generation in the second half of the 21st century. This will lead to increased holding of tritium in the fuel cycle, increased occupational exposure (including to tritiated particles) and increasing accumulations of tritium contaminated wastes. In this context, the scientific basis underpinning radiation protection in respect of tritium should be further enhanced, in particular the quantitative assessment of the transformation of HTO into OBT (in particular into DNA and histone precursors) by organisms and the transfer of these compounds through the food chain to humans. Particular attention should be given to establishing direct human evidence of the risks of exposure to tritium from epidemiological studies of appropriate cohorts. Those exposed in the production and use of tritium in defence activities and in the operation of heavy water reactors offer considerable potential in this respect.