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ORIGINAL PAPER

The hazards of tritium – revisited

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Tritium (^{3}H) is the radioactive isotope of hydrogen, with a half-life of 12.3 years. It is created naturally in the atmosphere, and in higher annual rates in nuclear reactors and in nuclear weapon tests. This article surveys the properties of tritium, its biokinetics and its biological effectiveness. The safety levels of tritium have been a subject of dispute for many years, as many scientists consider that its doses and risks, as promulgated by the International Commission on Radiological Protection are, too low and should be at least doubled. Recent reports and evidence of increased cancer risks near nuclear installations that release tritium are discussed; these are of interest in view of new proposals to expand civil nuclear power.

Keywords: β-radiation; internal emitters; International Commission on Radiation Protection; organically-bound tritium; radiation epidemiology; radiation protection; tritium

Introduction

A report on tritium by the United Kingdom government's senior radiation committee[1], together with a flurry of other tritium reports, have revealed renewed interest in this radionuclide within the scientific community. This article, therefore, revisits tritium and reviews these reports, examining in particular their implications for radiation protection.

Tritium (^{3}H) is the radioactive isotope of hydrogen. It is a low-range β-emitter with a half-life of 12.3 years and a maximum decay energy of 18.2 keV (average 5.7 keV). Tritium is formed naturally through cosmic ray interactions in the upper atmosphere, though anthropogenic tritium emission rates considerably exceed its natural production rate. Tritium most commonly occurs as tritiated water (^{3}HOH), and in some industrial/military instances as elemental tritium gas (^{3}HH), which is steadily oxidized to ^{3}HOH in the environment. Therefore, in most instances, tritium can be accurately described as radioactive water. Tritium is created in most nuclear reactors by activation of hydrogen (^{1}H) in their cooling water and

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moderator circuits and as a tertiary fission product in nuclear fuel. In heavy water reactors, larger amounts of tritium are created by the quicker activation of deuterium (^{2}H) in the heavy water of their cooling and moderator circuits.

Because of the low range of its β particles, radiation exposures from tritium only occur when it is inside the body – that is, tritium is considered an internal emitter. This does not mean that tritium outside the body is harmless, as tritiated water vapour readily permeates the skin and, when inhaled, easily transfers across lung and buccal membranes.

Main sources

Tritium is by far the most common nuclide encountered in radiation protection as it is emitted in large quantities from all nuclear facilities – both military and civil. On the military side, the largest sources are military production reactors and nuclear weapons manufacturing facilities, as tritium is used in nuclear weapons as trigger and reflector. These include the very large United States facilities at Savannah River, Hanford, Rocky Flats, Fernald and Oak Ridge, and the similar-sized Russian facilities at Chelyabinsk, Tomsk and Krasnoyarsk. Data from these facilities is sparse and only available up to 1982. For unknown reasons, the normally comprehensive United Nations Scientific Committee on the Effects of Atomic Radiation reports do not contain data on tritium releases from these facilities after 1982. Another military source is nuclear submarines, which discharge tritium from their reactors during refitting in port (for example at Devonport, UK).

Tritium is also created in nuclear detonations at the rate of 740 PBq per megaton (1 petabecquerel = 10^{15} becquerels – an extremely large amount of radioactivity). As a result, 186,000 PBq of tritium were released from the atmospheric bomb tests in the 1950s and 1960s and distributed throughout the world[2]. About 95% of this has decayed. Table 1 shows annual tritium production rates from major sources. It can be seen that very large amounts of tritium have been released in the past, especially from nuclear weapons production facilities.

On the civil side, tritium is the largest of the nuclide emissions from all nuclear reactors, apart from noble gases in some types of reactors. The highest emissions are from heavy water reactors (such as Candu reactors in Canada), from nuclear reprocessing plants (for example at Sellafield in the UK and La Hague in France), and from pressurized water and boiling water reactors. Another major source of tritium emissions is isotope manufacturing facilities. Tritium is also widely used as a tracer in medical research and industrial laboratories and various industrial processes, and as an energy source in emergency lighting equipment. Perhaps unsurprisingly, it is the main nuclide found in leachates from landfill waste sites in the UK. Table 2 sets out

Table 1. Tritium production rates of major sources.

Source	Annual emissions*	Total*	Source ref.
Nuclear weapons manufacturing (to 1982)	28,000	>300,000 (estimate)	3
Atmospheric weapons testing 1954–2000	–	186,000 (to 2000)	2
Natural formation in atmosphere	72	–	2
Civil nuclear power 1995–1997	28 (annual average)	300 (to 1997)	2 (Table 43, Annex C)
Emergency lighting (SRB Technology, Canada)	0.2–17	88 (1996 to 2006)	4
Fusion (estimate for future 1 GW facility)	0.11 (37,000 in accident)	–	5

*Emissions in petabecquerels per annum (1 PBq = 10^{15} Bq).

Table 2. Principal tritium releases from UK establishments in 2006.

Establishment	Emissions to air*	Discharges to sea*
Cardiff – Amersham plc	319	24.8
Sellafield – Nuclear Decommissioning Agency	187	1090
Chapelcross – MoD tritium production	121	0.1
Winfrith – Atomic energy authority technology	10.1	16
Hinkley point B advanced gas-cooled reactor	6.5	309
Wylfa magnox reactor	2.7	3.3
Sizewell B pressurized water reactor	1.2	55.1
Aldermaston – MoD	1.2	0.001 to Thames river
Dounreay – UK Atomic Energy Authority	0.3	0.3

Source: Ref. 6.
*Emissions in terabecquerels per annum (1 TBq = 10^{12} Bq).

recent tritium releases from UK facilities. Currently, the largest UK tritium releases to air (more significant for human exposures than discharges to sea) are from the Amersham plc facilities at Cardiff in Wales which manufactures radiopharmaceuticals.

Finally, tritium would be released in large quantities from any commercial fusion facilities in the future[5]. In the case of fusion accidents or fires, it is estimated that extremely large quantities of tritium would be released[7]. These estimates run contrary to the widespread, but erroneous, view that fusion energy is free from radioactivity; clearly, the opposite is the case[8,9].

A misunderstood nuclide

Tritium is also the most studied radionuclide: since the 1950s several hundreds, perhaps thousands, of scientific articles have examined its biological effectiveness (that is, its hazard) and other properties. Yet it remains a misunderstood nuclide, as some radiation protection scientists still consider it a 'weak' nuclide, incorrectly thinking that, as its β particle has low energy, therefore its exposures are of little consequence and tritium outside the body is harmless.

These are major misconceptions. In radiobiology, so-called 'weak' particles in fact have higher radiobiological effectiveness than more powerful emitters. Paradoxically, the lower their energy, the more effective they become. For example, β particles from tritium are actually two to three times more damaging than γ rays (explained later). Therefore to describe β particles from tritium as 'weak' is misleading: it is better to term them 'low-range'. The reason for the greater effectiveness of low range particles has to

do with the track structure of ionizing radiations. So-called 'strong' radiations (such as γ rays from cobalt-60) have very long tracks in tissue, but most of their energy is frittered away in small amounts over their long tracks. Damaging amounts of energy are deposited only at the ends of tracks. Low-range β emitters such as tritium effectively consist *only* of such track ends, and therefore are more damaging per disintegration than higher energy emitters.

Unfortunately, the International Commission on Radiological Protection (ICRP) still recommends that radiation from tritium is not particularly dangerous in comparison with other kinds of radiation. However, recent reports show a widening recognition that tritium is more hazardous than presently acknowledged by the ICRP; with the only question remaining being when the ICRP will acknowledge these reports. Unfortunately, the ICRP continues to ignore the copious available scientific evidence on the added hazards of tritium.

Properties of tritium

In many respects, tritium has characteristics marking it out as an unusually hazardous radionuclide. These include its extreme mobility and cycling in the biosphere, its multiple pathways to man, its instantaneous ability to swap with H atoms in all other materials; its comparatively high relative biological effectiveness (RBE), its binding with cell constituents to form organically-bound tritium (OBT), and the heterogeneous distribution of OBT in humans. More generally:

> Tritium has certain characteristics that present unique challenges for dosimetry and health-risk assessment. For example, in the gas form, tritium can diffuse through almost any container, including those made of steel, aluminium and plastic. In the oxide form, tritium can generally not be detected by commonly used survey instruments. In the environment, tritium can be taken up by all hydrogen-containing molecules[10].

Tritium emitted as water vapour or discharged as water from various facilities travels rapidly through multiple environmental pathways as water to reach humans, and cycles in the biosphere. Tritium atoms exchange very quickly with stable hydrogen atoms in the biosphere and hydrosphere downwind of a facility. This means that open water surfaces and biota downwind, including food growing in the area and food in open-air markets, and humans themselves would quickly become contaminated by tritiated air moisture up to ambient levels – that is, to the tritium concentration in water vapour in the air.

Humans can become tritiated not only by skin absorption but by inhalation of contaminated water vapour, and by ingestion of contaminated food and water. When tritium enters the body, it is readily taken up and

used in metabolic reactions and in cellular growth: over 60% of the body's atoms are hydrogen atoms and every day about 5% of these are engaged in metabolic reactions and cell proliferation. The result is that a proportion of the tritium taken in is fixed to proteins, lipids and carbohydrates, and most importantly to nucleoproteins such as DNA. This is called organically-bound tritium (OBT), which is non-uniformly distributed in the body and which is retained for longer periods than tritiated water (HTO). (All ICRP dosimetric models assume the opposite – that nuclides are homogenously distributed in the body/tissue organ of interest). Doses from OBT are therefore higher than from HTO. The longer people are exposed to tritiated water, the higher their levels of OBT become until, in the case of very lengthy exposures lasting for years, equilibrium is established.

Tritium, therefore, has unusual and noteworthy properties, which suggests that it would be noted as hazardous in radiation protection advice. Unfortunately, these properties are not recognized by the ICRP and by those radiation protection authorities which take their lead from the ICRP. This bad situation is made worse by the ICRP's incorrect dose models for tritium, which result in underestimates of 'doses' from tritium and its risks (explained later). The controversy is over the 'effectiveness' – that is, hazard – of tritium as interpreted by the ICRP and it has lasted more than 40 years (a future MCS article will discuss this matter). It should be borne in mind that the ICRP is not an official, but a voluntary, body. On occasion, it can appear to be more concerned with commercial or political interests rather than with the protection of the public. It appears that non-scientific considerations may have played a part in the ICRP's decisions on tritium, as regards nuclear weapons production plants in the past and fusion facilities more recently.

Hazard index of radionuclides

This raises the question about how radiation protection authorities classify the potentially hazardous nature of radionuclides. The short answer is that they do not: there is no comprehensive hazard index for radionuclides as there is for chemicals. Many scientists consider there should be one because the properties of nuclides would be better recognized if such an index existed. After all, some nuclides are considered much more potent than others (polonium-210, for example, used recently allegedly to poison the Russian dissident, Alexander Litvinenko). It has been suggested that a number of characteristics should be included in a hazard index[11]:

- large releases to environment;
- widely used in society (industrial/military/research/medical uses);
- rapid nuclide transport, solubility and cycling in biosphere;
- global distribution and resulting large collective doses;

- many environmental pathways to humans;
- rapid molecular exchange rates (that is, fast uptake by humans);
- large uptake fractions to blood after intake;
- organic binding in biota;
- long biological half-life in humans;
- long radiological half-life;
- long nuclide decay chains with radiotoxic daughters;
- high radiotoxicity (the dose coefficient of the nuclide, that is, the radiation dose imparted from the disintegration of one atom of the nuclide in question).

Tritium is unique in that it exhibits so many of these characteristics – in fact, ten of the above twelve, with most other nuclides exhibiting two or perhaps three traits. Polonium-210 has four, carbon-14, iodine-129 and krypton-85 have six or seven out of the twelve traits. But, as stated above, no hazard index exists for radionuclides – at least at present. It is recommended that national radiation protection authorities should take steps to set up such a hazard index.

This raises a further question – just how do radiation authorities gauge the relative hazards of nuclides at present? The answer is by estimating radiation 'dose' from the nuclide to an exposed person from one disintegration of that nuclide. This is discussed in Box 1 below, but using 'dose' alone ignores the first six of the above twelve characteristics. In other words, 'dose' by itself is an inadequate indicator of hazard for some important radionuclides, and for tritium 'dose' is a very poor one.

Recent reports

As stated earlier, a number of reports have recently examined the dosimetry of tritium. All of these reviews recommend that the doses of tritium should be increased by factors of two to five[4,12–15]. Significantly, the US Government's Environmental Protection Agency (EPA) recommended a 2.5-fold increase[13].

But the story really belongs in the UK where the tritium issue has been studied most consistently at least since 2000. The UK Government's CERRIE Committee on internal emitters[16] examined tritium in considerable detail between 2001 and 2004. Tritium is an example, *par excellence*, of an internal emitter, so it was no surprise that the Committee examined the nuclide in some depth. The report discussed a number of scientific arguments for increasing the dose coefficient of tritium by a factor of ten or more, but the Committee could not reach a consensus and in the end made no recommendation on increasing its dose coefficient. It is noted that three senior members of ICRP committees sat on the CERRIE Committee. They remained adamantly opposed to any proposed increase in the

Box 1.

How is 'dose' estimated?
In very simple terms, the amounts of a radionuclide inside a person are estimated by *biokinetic models*. These amounts are then multiplied by the radiation from the decay of the nuclide which is estimated by *dosimetric models*. The product of these two is the 'dose', discussed in the CERRIE report[16].

One problem is that these models used are only as good as their methodologies and their assumptions, and there are many questions about these. A second related problem is that there are many uncertainties in the estimated doses arrived at by these models, discussed at some length in the CERRIE report, which concluded that for some nuclides the uncertainties could be very large.

An important parameter used in dosimetric models is the 'dose coefficient' for each radionuclide (one for ingestion and one for inhalation). This estimates the amount of radiation emitted from the intake of an atom of a radionuclide. It is based partly on the biokinetics of the chemical form of the nuclide and partly on radiation physics. This dose coefficient is the last characteristic in the list of hazardous properties discussed in the text.

However, paradoxically, tritium has the lowest ICRP dose coefficient of all radionuclides by a considerable margin. For example, the ingestion dose coefficient of tritium is 30 times lower than that for carbon-14 (which is similar to tritium in some respects) and 660 times lower that for caesium-137. Thus, the hazards of tritium may be inadequately recognized by the ICRP, and the dose coefficient of tritium may be considerably greater than the current value estimated by the ICRP.

radiobiological effectiveness of tritium. It is of interest to note that their main defence – a Canadian study on carcinogenesis in mice exposed to tritium – was comprehensively demolished in the later Advisory Group on Ionizing Radiation report (explained later). However the tritium issue was picked up by the UK Government's permanent Committee on the Medical Aspects of Radiation in the Environment, which in 2005 referred the matter to the UK Government's senior committee on ionizing radiation, the AGIR, which in turn published its report at the end of 2007[17].

The AGIR report, at 90 pages, is by far the most comprehensive of the various studies and a close perusal is recommended. The main areas covered are:

- the properties of tritium;
- the RBE and radiation weighting factor (w_R) of tritium;

- biokinetic models for tritium;
- epidemiology.

Biological effectiveness

The longest chapter of the AGIR report thoroughly investigates the RBE of tritium and the report is to be welcomed for this alone. It reveals extensive radiobiological evidence (from cell and animal studies) that the RBE (that is, its hazardous nature relative to γ rays) is between 1.5 and three with an average of about 2.5. The report finds that the RBE value for tritium lies between two and three, but states that a value of two is '... most appropriate, based largely on an analysis of the available experimental data with rounding and biophysical considerations ...' and adds 'fractional values were not considered appropriate'. This conclusion can be criticized, as two is certainly not a precautionary value; three would have been the safer choice. In addition, no reason is given for not adopting a value of 2.5, which is directly supported by much experimental data. The report hints that fractional values were not used because of implied but spurious precision. But in fact the experimental data is copious and would have permitted the use of two significant figures, so that the more accurate value of 2.5 could have been chosen (and was in fact chosen by the US EPA[13]).

RBE values are used for specialist purposes, such as estimating doses in specific areas, for example, radiation biology studies. More important is the radiation weighting factor w_R, as this is used for general radiation protection purposes, such as legal authorizations for nuclide discharges from industrial facilities. In a rational world, the w_R value for a particular radiation should be *more* protective (that is, larger) than its corresponding RBE value, but for tritium the opposite is the case. An increasing number of scientists use values of two to three for the RBE of tritium for experimental purposes, but for *radiation protection purposes*, the ICRP, bizarrely, recommends a *less* safe value of one. This situation should be rectified as soon as possible by national radiation protection authorities.

The AGIR report concluded that: '... consideration be given to the use of a value of two for radiation w_R in routine radiation protection assessments for tritium' (page 3 of the Executive Summary). This is by far the most important recommendation in its report, and the AGIR are to be congratulated for making it, faced with the ICRP's reluctance towards making any such statement.

Biokinetic models

The ICRP's biokinetic models for tritium are questionable for a number of reasons. First, the ICRP's values for important parameters in these models

(the percentage of activity taken from blood into the tissues and the biological half-life) are poorly supported by the majority of available animal and human data. Second, the ICRP only models single intakes, not protracted ones, although the latter are much more common in the environment, for example, intakes by residents near industrial facilities. ICRP representatives have argued that a chronic exposure is merely the sum of single exposures, but this is clearly wrong. For instance, from each single administration of HTO the dose from OBT is neglected, but OBT doses from chronic exposures are significant. Third, the ICRP models all assume that most of the committed dose from tritiated water intakes is from HTO, when much evidence indicates that, after the cessation of intake, cumulative dose will continue to rise mostly from OBT, so that in the longer term, OBT doses are greater than HTO doses.

Unfortunately the chapter on biokinetic models in the AGIR report fails to discuss these matters clearly. It states that, as a result of chronic intakes of tritiated water '... equilibrium amounts of OBT and HTO will be established in the body', and there is certainly an abundance of data (unfortunately not cited in the report) indicating that this occurs in actual practice. But the chapter fails to discuss the fact that ICRP models do not take this crucial matter into account when assessing tritium doses.

Even more perplexing, the chapter frequently asserts that human and animal data provide general support for the ICRP's models. But this is incorrect, as the evidence cited in the chapter[18,19] contradicts the ICRP models, and reveals that the parameters used in the ICRP models are not at all precautionary. A key point is that even if one were to use the ICRP's unsafe parameters, it can be shown that, at equilibrium after chronic exposures to HTO, OBT doses would be about four times greater than those estimated from an acute exposure. The implications of this for estimates of doses from tritium are ignored in the ICRP's models and in the AGIR chapter as well.

Furthermore, it is disturbing that a great deal of radiobiological evidence on tritium uptakes and retention in animals has simply been ignored. A good account of the correct biokinetic models to be used in the case of tritium therefore remains to be written. This is an important matter as these models directly impinge on the assessment of doses from tritium. For the time being, I have suggested elsewhere that the dose coefficient of tritium should be increased at least by the above factor of four to take these matters into account[14].

Epidemiology

Unfortunately no epidemiological studies directly assess the effects of tritium exposures alone[20]. However, there is indicative evidence suggesting increased incidences of childhood leukaemias and congenital malformations

in populations exposed to tritium as well as other forms of radiation. Indeed there is important new epidemiological evidence of increased leukaemia incidences near German nuclear reactors (the KiKK studies)[21]; these reactors emit relatively large amounts of tritium.

Perhaps the most arresting evidence in the AGIR report was a study[22], which examined mortality in more than 45,000 Canadian nuclear workers between 1957 and 1994. Tritium doses were calculated from urinalysis data, and added to external (film-badge) doses. The study did not indicate tritium doses but stated that, for some workers, these could have been large. Overall, the mean dose among those having 'some dose' was 19.7 mSv. The resulting excess relative risks were very high and were mostly statistically significant, though with wide confidence intervals, as shown in Table 3.

Similar cancer epidemiological studies in the past have often indicated small increases in cancer risks over the spontaneous risk – such as relative risks of between 1 and 2 to 3 per Sv. However, Table 3 indicates that the Canadian workers' background leukaemia risk was increased more than 50-fold per Sv of exposure, but there is quite a bit of uncertainty in the actual size of the risk. The study thus provides indicative, but not conclusive, evidence of the magnitude of the increased risks.

Nevertheless, if we were to apply the observed excess relative risk (ERR) of 52.5 per Sv for leukaemia to those nuclear workers whose average dose was 20 mSv, their average excess risk would be 1.05, that is, their background leukaemia risk would be more than doubled. This should be considered along with the fact that the spontaneous leukaemia risk in most Western populations is very small – causing fewer than two deaths per 1000 in the UK, for example.

Table 3. Radiation risks in Canadian nuclear workers.

	Excess relative risk per sievert	95% Confidence intervals	2-sided p test
All leukaemia excluding CLL	52.5	0.205, 291	0.048*
Rectal cancer	34.1	1.41, 165	0.029*
All solid cancers	2.80	−0.038, 7.13	0.054+

Source: Ref. 22.

*Statistically significant at 5% level.

+ Borderline statistical significance.

CLL: Chronic lymphocytic leukaemia, which is not thought to be due to radiation exposures.

Relative risk: the cancer risk expressed as a multiple of the background cancer risk. A relative risk of 3 per sievert means that an exposure to 1 Sv would treble your risk of cancer. Excess relative risk = relative risk − 1. In the example above, a RR of 3 is the same as a ERR of 3 − 1 = 2.

95% confidence intervals are two numbers within which we are 95% sure the true value lies. In the first row of the table we can be 95% sure that the correct value is between 0.2 and 290.

Similarly, the ERR for all solid cancers of 2.80 per Sv means that the nuclear workers' excess risk would be 0.056 (a 5 to 6% increase in the spontaneous risk). But the background risk of solid cancers in Western countries is very large, about 25% of all deaths in the UK. So the picture shown by this study is of a big increase in the small spontaneous risk of leukaemia, and a small increase in the large spontaneous risk of solid cancer.

What is clearly required are more epidemiology studies, and the AGIR report recommends that immediate meta-studies be commissioned to reconstruct tritium doses in previous epidemiological studies in order to provide harder information on tritium risks. This is certainly to be welcomed. It is understood that these studies are being established in some countries including Canada.

Conclusions

In sum, the AGIR report is mixed: most of the chapters are very good and will amply repay study, other chapters less so.

The most important conclusion of the report is that official consideration be given to the value of two for the radiation w_R of tritium in routine radiation protection assessments. This was directly aimed at the ICRP, as is clear from the remarks by the AGIR Chairman, Professor Bryn Bridges[23] when he stated, 'A lot of work went into this report and I hope the International Commission on Radiological Protection will consider our suggestion.' However, the ICRP has recently announced it will not do so, citing spurious non-scientific reasons such as alleged uncertainty and convenience[24,25].

It is likely that the ICRP will be criticized for its failure to recognize the overwhelming scientific evidence on the hazard of tritium and for the possible non-scientific reasons for its decision. But perhaps its views may be irrelevant in the end. In recent years, the reputation of the ICRP has declined as a result of external criticisms of its conservative attitudes, and the critical responses (even by the nuclear industry) to its 2004 draft recommendations on radiation protection, which were effectively withdrawn as a result. Already a number of researchers are using RBE values of two or more for tritium in certain areas in defiance of the ICRP's current recommendation.

Instead, it is likely that UK and US government bodies responsible for radiation protection will now use a radiation w_R of two for tritium regardless of the ICRP's attitudes, and will require the UK and US nuclear industries and others to follow suit when estimating doses. The same is likely of the European Commission and other European governments and their agencies; if these agencies were to hesitate to change, they would open themselves to challenges from members of their publics pointing to the safer recommendations in the official UK Government report.

Overall, it is concluded that major rethinks are required on tritium risks and tritium 'doses' among the radiation protection community. The comprehensive AGIR review, recent US EPA report and the reports of increased leukaemia risks near German nuclear reactors discussed above together provide much food for thought on tritium's dosimetry and its risks.

Notes on contributor

Ian Fairlie is an independent consultant on radioactivity in the environment. He has degrees in chemistry and radiation biology, and his doctoral studies at Imperial College examined the radiological impacts of reprocessing discharges at Sellafield and Cap de la Hague. He has worked for various UK government departments and regulatory agencies, and advises environmental NGOs, the European Parliament, and local authorities. Between 2001 and 2004 he was Secretariat to the UK government's CERRIE committee.

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