A Story of T

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Unfortunately, Dr. Richard Osborne passed away before he was able to complete his Sievert Lecture paper. In its place is this very lightly edited transcript of his Sievert Lecture, presented at the 13th International Congress of the International Radiation Protection Association in Glasgow, UK, May 14, 2012.

Let me tell you a story about tritium. It's really several short stories. You might well ask, why tritium? In many countries, there's a continuing public interest in exposure to tritium, and in that sense it's very complimentary to the conference theme. Most of my research and development work at Chalk River Nuclear Laboratories has been related to tritium, so it is a subject that I know a little about, which is always a good criterion for any lecture. These stories will illustrate the wide range of disciplines in radiological protection and I'm hoping for those newcomers to the profession some insight into the really exciting wide range of disciplines we get involved in. There are certainly areas related to tritium where research is needed, and I'll try and highlight those as I go through, and some of these issues have a boarder application in radiological protection, so that's why tritium.

So what I'm going to do is to give a quick overview, talk about the early days and then go through the various topics which illustrate the wide range of disciplines of measurement, biokinetics and dosimetry, relative biological effectiveness (RBE), dispersion in the environment, health effects, effluent management and then a summary. So you see it's a series of short stories, not just one story.
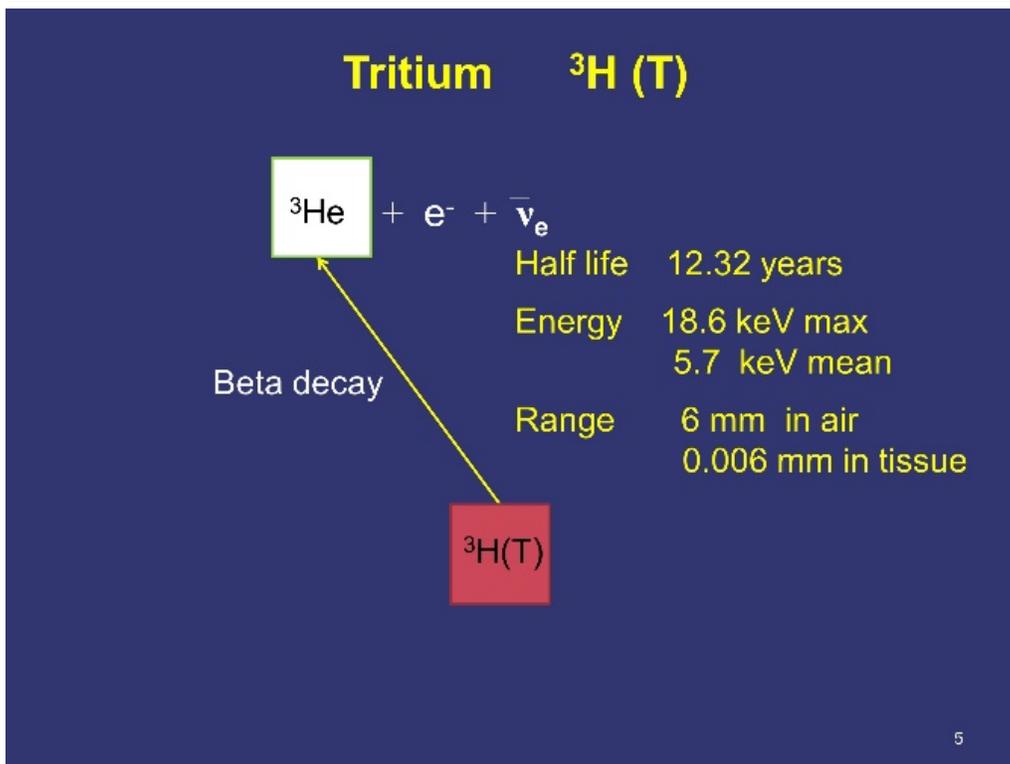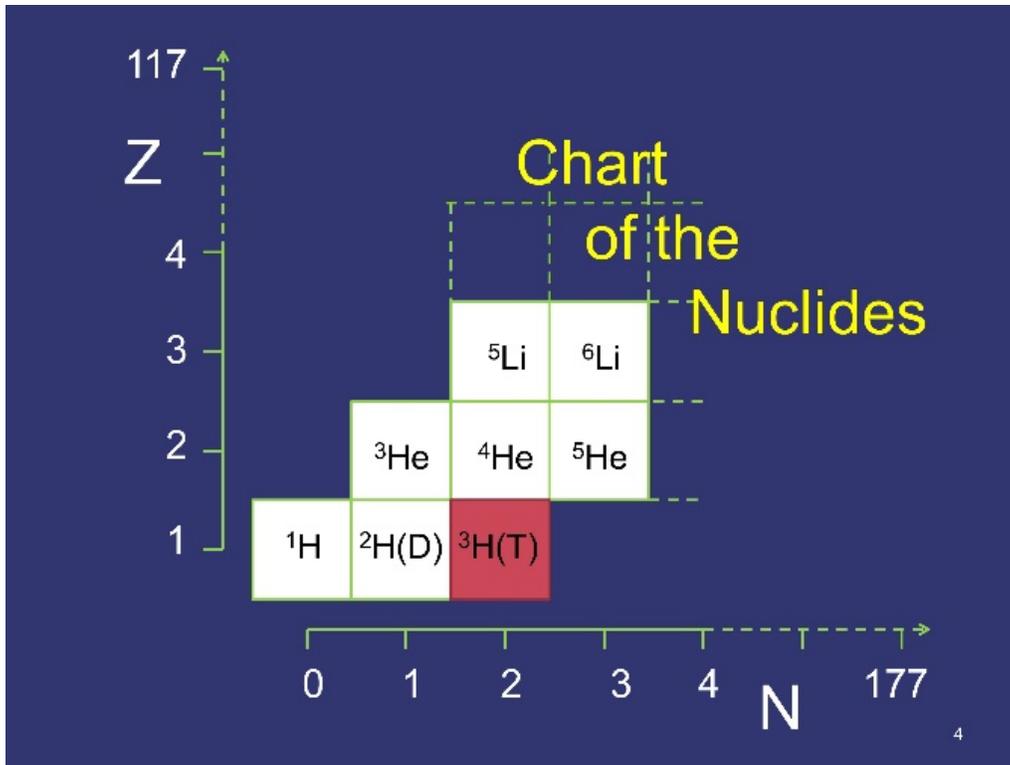
Figure 1. Tritium on the chart of nuclides (above), and its decay scheme (below)

Let's take a brief look at tritium. If we look down at the bottom end of the chart of the nuclides (Figure 1), we find the three isotopes of hydrogen, and we see H-3, the radioactive isotope of hydrogen known as tritium, decays to He-3 with an anti-neutrino. An important parameter here is the half-life of tritium of 12 or so years, which means once it gets in the environment it tends to stay there for decades. The energy is relatively low, the maximum energy is 18.6 keV, and the range consequently is fairly short in air and even shorter in tissue. This has two main implications, one is it makes it quite hard to measure and secondly it means it's only what we would call it internally matter of radiological consequence, radiation from external from the body, really isn't in great concern; is only when you take the tritium in, is a concern. It's produced in a variety of ways, naturally produced tritium produced by cosmic rays or O-16 and N-14, it's a tertiary fission product in nuclear reactors and weapons. We find neutron capture from by deuterium in heavy water reactors, and also in the (n-p) reaction on H-3 which is formed from tritium and obviously this is important if the heavy water is not purged. It is also formed by neutron capture on Li-6 in other reactors. It's got a number of uses which we know for well, nuclear fusion research in the third world nuclear weapons, it's used extensively in biochemical and hydrological research, and it is used in light sources. Into the environment the natural production produces about 72 PBq y^{-1}, and this results in a few fractions or a few Bq L^{-1} in moisture which is naturally produced. In nuclear reactors, from fission what is released to the environment – and this is of a few years ago and of course this varies from year to year – about 13 PBq y^{-1}. And for weapons and thermal nuclear weapons, before the Test Ban Treaty came into place in the 1950s and the early 1960s, injected into the atmosphere was about 186,000 PBq/y. But consequence of this is in the environment we find tritium in three main chemical forms: high tritiated hydrogen (HT), tritiated water (HTO) and tritium bound in carbon compounds which we call organically bound tritium (OBT).

Going back to the early days in the mid-1930s, Rutherford was playing around in the cabin dish with deuterium and deuterons, and was the first one to observe these heavy hydrogen isotopes. He wrote – and this was in the Nature article – "diplons have been used to bombard preparations in which the hydrogen has been displaced in large part by diplogen", we call it deuterium and deuterons now of course. He concluded, "While the nuclei of H-3 and He-3 appear stable for the short time required for their detection, the question of their permanence requires further consideration". So he knew he got something, he wasn't quite sure what he got. Uncharacteristically of Rutherford, he made a mistake. He thought that H-3 (what we call tritium) would be the stable compound, and asked Aston who had just invented the mass-spectrometer to look for it. Not surprisingly, Aston couldn't find it, and in fact, it was left to Alvarez in the U.S. to

actually detect tritium with such, and he wrote in Physical Review, "Since we have shown that He-3 is stable, it seemed worthwhile to search for the radioactivity of H-3", and he concluded "The radiation emitted by this hydrogen is of very short range". So, for the first time, somebody had seen tritium and done a something about it.

Well after the Second World War and the late 1940s and 1950s, natural tritium was detected by Faltings and Harteck in Germany, and it became used extensively as a tracer for atmospheric circulation patterns and in hydrology. People were drawn to the fact that this was a very useful tracer. But in 1950s and 1960s, concerns started to be raised; this shows the increase in tritium in moisture around the world as a result of weapons testing. You see the various events through the years, collating in the very large 63 events, with tritium concentrations and moisture getting up to around six or seven hundred Bq L^{-1}. This was measured at the Ottawa Station, there were stations measured all around the world, and some particularly in northern Canada was much higher than this.

Also in the 1950s and 1960s, we started to get some concerns about occupational doses from tritium. In Savannah River in the U.S., a series of heavy water reactors started to build, these were heavy water moderated and cooled. In Canada, at Chalk River, two reactors were built, in 1947 – the NRX reactor, and in 1957 – the NRU reactor; and you can see that these two made bigger buildings in the picture there. In fact, it looks a little better in colour in somewhat later photograph, I always have to explain at this stage that the colours in the foliage have nothing to do with radiation from tritium, it's just part of the glorious Canadian fall colours that we see. I also want to point out that NRU is still running after it started in 1957 and still producing a lot of the world's medical radioisotopes, which is quite an achievement. When I arrived there in the early 1960s, the workplace concerns were with measurement and monitoring of tritium, with skin absorption and with dosimetry, and I'll talk a little about those experiments that we did and the conclusions we came to.

So first of all measurement, I'll restrict myself to measurements in air, just because we also measure in water, in automatic urine analysis, but I'll just stick to air monitoring needs. The first need was to have something practical, and I go back to my earlier mentor, who used to stand in front of me whack his finger and say, "Richard, handsome is as handsome does", quoting Chaucer, when I would explain how wonderful something I had invented was going to work. And that's very true when you get into designing instruments to work in reactor environments; you really have to make sure they are rugged and practical; you need adequate sensitivity and we have the constraint

of the short range of the tritium beater; you need to discriminate against gamma background and also against radioactive noble gases that you find in the reactor environments. So, there's a lot of development in the late 1950s through into the 1980s in the research and development laboratories at Chalk River, in the States at Los Alamos, Livermore, Savannah River, in Germany, and these developments for tritium monitoring, I say provided the basis for the methods you find currently used. I was somewhat surprised when I compared the text and the report we wrote in 1976 for National Council on Radiation Protection and Measurements (NCRP) with a review my colleague Mike did in 1993 and a more recent review Marsh did recently for his thesis in Southampton just a couple of years ago. The techniques were much the same, the sensitivities were much the same.

I'm just going to run through some of those particular techniques. First of all, for detecting tritium in the gaseous phase, the way you do it really is a flow-through detector; you have to get tritium in close contact with a detector or within it. The ionization chamber is by far the cost of this kind of detection. The downside is, the Bq of tritium only produces about 25 aA, which is kind of a small current. A derived air concentration (DAC) as it is called now, which I'm taking as 0.3 MBq m^{-3}; in a one litre ionization chamber, which gives you about 7.5 fA; so 40 litre ionization chamber gives you about 0.3 pA. In the 1960s, we could manage to measure those sorts of currents. The problem was that a very small gamma field, 0.8 µSv h^{-1}, would produce about the same current. So, it was kind of swamp tritium measurements, the way around this is to use two ionization chambers, one sealed, the other that sample the tritium. We tried various combinations of ionization chambers, and decided that the concentric of arrangements with a sealed chamber inside an outer one that was ventilated, was by far the most effective way of getting this compensation. I was amused actually to see a paper quite recently where somebody had done a Monte Carlo calculation on the various arrangements of chambers and decided that yes indeed that was the best way of doing it. They concluded having seen our paper that we must have done our experiments correctly. Now, as an experimentalist, was I think they must have done their calculations right. Anyways, that is the manifestation of one these 40 litre chambers, and interestingly George Cowper and I described this particular instrument at the 1st International Radiation Protection Association (IRPA) Congress in 1966; well that in fact gives you about 98% gamma compensation. I'm rather amused to find – when I looked in NRU just a couple months ago – some of those instruments were still in operation 50 years later with a slightly electronics, still looking rather old but still working very well and still relied on.

The problem with those, they do gamma very well but they still respond to noble gases. Ar-41 for example, the same concentration as tritium in air gives you about five times the ionization in this 40-litre volume. Simple way around this is to use a double set of chambers with a desiccant in between, and the net current then gives you the difference of the tritium signal, and you can get very good compensation for gamma and noble gases, at about 99% in this way; and we described this at the 4th fourth IRPA Congress.

The other way is to take the tritiated water which is formed in the atmosphere that we deal with, into the liquid phase. There are a number of ways in doing that, continuous exchange is one that we had explored using either a plastic or liquid scintillator to measure it. This is a continuous flow water exchanger that we designed. Sample air and water go in the top, the water picks up the HTO from the air, it flows on through, you purge out the noble gases, lower down the water flows onto a plastic scintillation detector. This gives you quite a good discrimination against noble gases, it will detect down to about a tenth of a DAC. I should make a side point here, one of the problems we had in the 1960s and 70s was that – it wasn't like present day when there are so many ways, like electronics and computers – we had to design much of our own electronics. For example, for this instrument, I designed a digital four-decade rate-metre, using what were new solid-state devices coming out on the market then. One of the points I would make here is that, many of these old designs are well worthwhile for current designers to back to them and see how much better they would be, now with the electronics that are available.

Going on to the last one of these, you can obviously get a very sensitive detection for tritium by doing the liquid exchange scintillator. There's a very neat material here called Nafion, which allows water molecules to go through and not much else, it's just the right size and it has a polar structure. So, with a count to count flow with air and HTO flying in, scintillator going through the centre, you can pick up the HTO will move into the scintillator, and you can then go into a standard liquid scintillation detector. This gives an extremely good discrimination against noble gases. Interestingly hydrogen doesn't go through the membrane, so you get a very good discrimination again HT as well if you are looking to make that discrimination.

The final method I will just briefly describe is one that made an enormous difference to our sampling, both in the workplace and in the environment, and that's what I call a diffuser sampling system. This is nothing more than a 20 mL liquid scintillation vial, it's got a small tube in the top, and by sizing it appropriately you can arrange that the vial with any fact sample, let's say 1 or 5 L of air d^{-1}, and any tritium that goes in is picked up

in the water or the water/glycol mix in the bottom. You can add a wet-proofed catalyst to it and convert tritiated hydrogen to tritiated water and collect it. So you can use it to measure both tritiated hydrogen and tritiated water in the atmosphere. This is a wonderful, doesn't require any power and you can set them out all over the place and sample for a week or longer.

So just quickly looking through those methods I described, we found the current instruments also can detect down to about 40,000 to 3000 Bq m^{-3} and if you recall the DAC is about 300,000 Bq m^{-3}, so it can detect down to a fraction of the DAC. The exchanger method is slightly more sensitive, but gives you better discrimination against noble gases. Bubblers or diffusers will get you down to a much lower concentration and I recall the natural levels in air would be something like 0.01 Bq m^{-3}, so on to that one with an appropriate liquid scintillation counter you can measure nature levels fairly easily. I just like to point which I find quite interesting, the most sensitive way of measuring tritium now is to use a mass spectrometer, and this of course is the instrument that Aston in the 1930s failed to measure tritium with. But the trick is not to try to measure the tritium itself but to measure the He-3 decay products that form in a sealed sample which you can leave sealed for week, or month, or even longer. And then count the He-3 atoms in the mass spectrometer. This gets you down to incredibly low concentrations.

I'll finish this section by saying that, the methods are there, they can be improved, one can get perhaps better discrimination; the ones with flow systems can probably take advantage of modern electronics, but by and large the systems are in place.

I'm going to shift tack now to biokinetics and dosimetry. There are a number of issues here: the intake through the skin, doses from OBT, tritium on surfaces, from tritiated particles, and a little comment about interpretation of bioassay results. I should point out that in Chalk River in 1949 there had been what is in essence the first international gathering on internal dosimetry – the first standard man parameters were fixed here. They actually made quite a decent stab at tritium dosimetry suggesting that 370 MBq would be the maximum body burden – the concept used then, then current limit of 3 mSv per week, which is actually not a bad estimate.

The first job I was given when I got to Chalk River was to find out how much tritium went in through the skin. At that time, Pinson and Langham in Los Alamos has done some measurements on the forearms of people came up with a value of 5 L min^{-1} m^{-2}. The value here on the ordinate is the air volume containing the tritium absorbed, so if you

multiply that by two, the area of the skin and the body is about 2 m^2, so multiplying it by two gives you the equivalent breathing rate to skin intake. So what I did, was with one measurement in Sweden, what I did was a series of measurements on 17 of my trusting colleagues, measuring tritium intake through the whole body and came up with a value of about 5 L min^{-1} m^{-2}, and that corresponds to about 9.7 L min^{-1} breathing. So if you are breathing in a tritium atmosphere, your breathing is about 10 L min^{-1}. The skin intake will just about double the total intake. I subsequently did some measurements from the forearm to look at the kinetics, I was quite surprised to find how well the skin behaves in a physical sense, so like a classical Fickian diffusion kinetics to follow, and to analysis the absorption curves, one comes up a delay time for the skin of about 10 minutes.

That was one sort of parameter settled. Just going to look now at what happens from the various intakes. Tritiated gas you might expect without a doubt, pretty much straight away; tritiated water will be eliminated in urine, breath moisture or perspiration. My colleague Peterman (Chalk River) showed that there was a small fraction of the HT converted to HTO, but the HTO was mostly breathed out fairly quickly. The tritiated water as we know for well has a half-life of about 10 days, lots of results have been reviewed by these two documents, as I noted there. I've seen half-life as short as three or four days, as long as 18 days, and certainly if someone goes the local pub and drinks beer every night, it gets your tritium half-life down very well, I think some work has probably done that. Some of the tritiated water is converted to OBT, and of course if you take in OBT then it is excreted in urine and faeces, but some are also converted to HTO. Tritiated particles may well be excreted in faeces, or they themselves tritium may be converted to organically bound and tritiated water. Tritium from the surfaces may end up as organically bound in the body or as tritiated water. These colour arrows I'm going to say a little more about because that's where we have some of the uncertainty in our parameters.

First of all, a dose from OBT after tritium intake, this is some work on Walter Snyder in Oak Ridge – an individual who received a fairly hefty dose of tritium was followed for a better part of the year. You can in a good mathematical way, put in a couple of straight lines in a semi-log plot and assign some sort of compartments to those body Walter's – the first one, and some organically bound – the second. You can do a slightly better job by using three fitted curves and you could actually do a good job too with the power law. It is tempting to try and assign some physiological quantity to these, but there really are only mathematical concepts, and you find people building up multi-compartment models. And I'm not – this is a fellow from Oak Ridge in 1982, I'm not picking on him –

but this is typical of many models, which really are mathematical concepts where you assign those particular exponentials to various compartments.

It is worthwhile stepping back and wondering about that, and considering J. von Neumann's adage, he said, "With four parameters I can fit an elephant… and with five I can make him wiggle his trunk". So as a cautionary word there, don't get carried away with all these parameters, I think it is useful to always go back to basics. If you consider brief exposure to tritium and think of an organic component A and all the organic components in the body are turned over in various time periods. If it takes a long time, a small portion of the components going to be lay over with tritium; if it's turned over fairly quickly, a larger portion is going to be labelled. If you go through the mathematics, you find that you really only need two parameters to estimate the dose from organically bound. The fraction of organically bound hydrogen labelled with tritium, which is about 20-30%; and the water fraction in tissue which is well 60-80%. And you end up with a dose from OBT of about 5-20% of the dose from HTO. Pinson and Langham were suggesting this and I further went into it in my radiation research paper in 1972. There has been some validation of that by my late colleague Trivedi, who measured tritium in the organically bound compound excreted in urine, and he estimated about 7% of the dose from OBT compared to that from tritiated water. The International Commission on Radiological Protection (ICRP) has a simple model, using – as I showed here – a 40-day half-life organically bound, and with that model, organically bound contributes about 10%. So, it's a reasonable model, it seems be in a with the experimental results. With that, the dose conversion coefficient for adults is about 18 pSv Bq^{-1}, and 64 pSv Bq^{-1} for an infant; the 18 are carried through with some of the next slides.

When it comes to the ingestion of OBT, it gets a little more complicated. There were some very early experimental studies by Lambert and Vennart in the UK, and they suggested the dose from OBT would be about three times higher than from tritiated water. There had been a number of subsequent studies suggesting values from one to four times higher. My colleague Richardson at Chalk River based it in a carbon model, suggested that dose would be about four times higher, so there is some variation there. ICRP currently take 50% of the OBT as being catabolized to HTO and they end up with a dose about 2.3 times higher. So in comparison, OBT gives a somewhat higher dose per unit intake than tritiated water.

Tritiated particles are not particularly a big problem in the nuclear industry so far. But when we come to fusion, then something to point out here, they are potential show-stoppers. What we are talking about here, are all the particles that we may end up with

which could be tritiated. We really don't have a lot of information on the behaviour of such particles, probably the best information is some work that Cheng did, reported over a decade ago on titanium hydroxide – this is the basis for the ICRP current model, which is used moderately for solubility for such particles and when you go through dosimetry you end up with a dose similar to OBT. The number that they have is actually 45 pSv Bq^{-1}. As a caveat on this, again my colleague from Chalk River, Richardson pointed out this may well be too high because it is ignoring self-absorption in the particles, it's ignoring microphage action and tritium speciation, so I think it serves to point out there's a fair bit of uncertainty to what the doses might well be from tritiated particles.

With tritium on surface or tritiated surfaces in which tritium gas is being absorbed, it's also uncertain. I tried and I found these experiments hard to do, it's very hard to get reproducible results. Eakins at Harwall in the 1970s did what I think is probably the best experiment so far. Certainly, HTO and OBT are formed in skin, a few percent of the tritium are transferred to the skin and then there's a slow release from the skin. But we can the dose from this or the effective dose is actually not very easy. John Johnson from Chalk River made one estimate, he suggested 10 pSv Bq^{-1}, but he agreed that it was pretty uncertain. So compared to the others, maybe 10 but it could well be less or it could be higher than that.

I just want to make one comment about bioassay, there's just one problem we have in bioassay is having measured tritium in urine deciding what the intake was. With particles and intake through the skin, it's really very complicated. Trivedi made some interesting observations some time ago, if you do liquid chromatography on the organic materials in urine, then the distribution of labelled compounds seems to be the characteristic of the type of intake, and this so far I could see hasn't been followed up. But that's something quite useful as we get into more concern about particles for example, in fusion systems. So I find those numbers are not as easy to remember, I always found useful talking to people in the public, whether it's a pound of tomatoes that's got so much tritium in it, that it's a real hazard. To point out that for an adult intake of 1 MBq of tritium, if it's tritiated water it's 20 µSv; it's three times that if it's organically bound; three times that for tritiated particles; for tritiated gas only about 1 in 10,000; and tritium on the surface, it may be about half way or it may be about the same. So, we need further experimental studies on OBT, on tritiated particles and surfaces, and interpretation of bioassay results.

Let me move on to the next chapter in this story, on RBE, which for tritium is a dose from reference radiation to produce a given effect, over the dose from tritium to produce the same effect. This is the ratio of course which influences how/what way you give to radiation doses in protection. It would be useful to have this value for chronic low doses which is what we are usually concerned with in protection. My guess from the variation in the different effectiveness is due mainly to the spatial distribution of energy disposition, and in the case of tritium we'd expect it to therefore be similar to 70 keV photons. So we expect it to be more like low energy x-rays perhaps than gamma rays. There have been some extensive reviews, by Little and Lambert and by the Canadian Nuclear Safety Commission (CNSC), and I'm not going to really deviate from their conclusions, I'm just pulling out some of the highlights. If we look at the results through the years, on measurements for RBE – this is for x-rays and for gamma – and you can say sure the gamma one looks a bit higher than the x-rays, and you might be tempted just to take a simple average. But really it isn't a good idea because there are for all sorts of different conditions, different endpoints, different organisms etc., so one has to be a bit more careful on that.

I would just like to point out that first by Furchner in Los Alamos that it was influential actually in the late 1950s. ICRP was persuaded actually to use a weighting factor – what we call a weighting factor or quality factor then – of 1.7 based on that particular result. But it was soon realized that it was not that relevant to radiological protection. It was the in mice, and they were shifted back to a unit weighting factor subsequently. What we really need from these results is the ones that are relevant to cancer, then we take out all the ones that are not cancer related and end up with just four. Lower two – the ones against x-rays, the ones carried out in my division in Chalk River. One on mammary tumours in rats, the other on leukaemia in mice. The reviews of all the studies had some criticism of them, they are hard experiments to do, and the reviewers have felt that the error bars probably didn't reflect the uncertainty in the results. But never the less, the average values were – as you might expect – about 1.2 for RBE against x-rays, and 2.5 against gamma. So it's nothing surprising here, tritium fits in where you might expect it to fit in.

So you then come to, "What is the choice of radiation weighting factor (w_R) for tritium?". And recall that, if you go from one end of the photon spectrum to the other you got a range of at least five in relative effectiveness, and we have an RBE for tritium that fits within that range for photons. So, it seems to me that ICRP's approach of using the single w_R value of 1 for all the photons is certainly applicable to tritium. It doesn't mean to say that if you are doing some actual risk estimate you might not use a slightly

different value. But of course given the uncertainties of those experiments that I talked about, it's probably worthwhile trying to make some definitive measurements for actual risk estimates, and I gathered in the biological research facility at Chalk River, some of those experiments are starting to take place with some international corporation.

Turning the page to my next chapter, a lot of other different disciplines now are talking about dispersion in the environment. We have a variety of models, as I'm sure you know for dispersion of HTO and HT. I'll just give some examples here, the Canadian ones primarily: accident releases from the Canadian Standards Association; chronic releases of HT and HTO from Peterson and Davis – of course this is interesting and little bit more as well; United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) values for regional and global dispersion. Now what these models are trying to stimulate are all these atmospheric processes. It is complicated, there are lots of parameters to measure and to validate, and I'm just going to talk about two of the more contentious ones perhaps. The conversion of HT to HTO in soils and the conversion to OBT in plants. There are two approaches to get a hang along the different parameters.

First of all, the HT to HTO, we have done a series of measurements at Chalk River, this just illustrates one of them. Where we released HT in the area around Chalk River into an experimental area and watch/see what happen to the tritium. Really, measuring the, what might be called deposition velocity for HTO from the HT, this shows some of the plants in the area was maybe an acre of two. Results gave some estimates of the parameters showing that maybe a percent or two of the tritiated gas was converted to HTO. This is obviously an application as parts of the database for testing short range HT dispersion models. We turn to HTO to organically bound conversions in animals, one can make use of the contaminated environment that we have. These are results from Canada, looking at tritium in moisture in various biological media: soil and vegetation and various food stuff; different distances from the nuclear generating stations in Ontario. As you might expect and remember – this is a log-log plot – as you move away from the stations the values go down. What's interesting is that when you measure – as was done – the ratio of the specific activities of tritium to hydrogen in the organic phase, to tritium to hydrogen in water. There is a fair variation, there's no distance correlation here; the average is about 1.3, where most is within a factor of two. But quite clearly, estimating any particular instance what the value is going to be, it's going to be very seasonal dependent or the other environmental variables.

If you take a typical fruit basket and use that average, I estimated maybe 13%-17% of the dose to the public would be from the OBT in food. There's certainly uncertainties though, it quite clearly points, there's a need to study the movements of OBT through the food chain and to make continuing improvements in models in tritium behaviour.

This brings me to what I think is an extremely important international program, this is the model intercomparisons and validation programs. I'm not sure if I can remember all these, BIOMOVS – biological measurements and validation study etc. The first one was started in 1985 in Sweden, we joined in very quickly as did the group in Spain. The subsequent programs have been run under the auspices of the International Atomic Energy Agency (IAEA) and this is extremely important. I'm just going to illustrate one from EMRAS, which took some Pickering counter cells. Pickering is one of the heavy water power reactors in Canada. And there were measurements of HTO in air, rain, soil, drinking water, plants, milk, and meat. There were measurements of OBT in plant and animal samples. The participants in this study – into comparison – are given measured concentrations of tritium in air, precipitation and drinking water. They were asked to calculate OBT and tritiated water in plants, milk, and meat and tritiated water in the top 5 cm of soil. I want to show the results given tritium in air, what the OBT would be in meat. These were the results, the green shows the measured value; the yellow was the various participant's estimates. They were mixed but some were pretty good, I think the important thing is not whether they were spot on, but whether the uncertainties they estimated they had included the real value, and it's also very important from a model's point of view, it then begins or helps you understand why you might have not got the right answer. So these sort of studies are really important as we go on to any models and try to improve them.

Let me move on to health effects. We know for well that there have been exposures of the public and to workers to tritium resulting from heavy water nuclear power plants and laboratories, nuclear fuel reprocessing, nuclear weapons development and production, fusion reactor research and development, and production of tritium sources for medical and industrial uses. You might well expect that one might be able to at least look for effects on health from here. Certainly there have been epidemiological studies of the public near nuclear facilities that release tritium and for nuclear workers exposed to tritium. I would expect you know there have been recent reviews from the Advisory Group on Ionising Radiation (AGIR) in U.K. and the CNSC in Canada, they have had extensive reviews of these studies. The significant effect observed is well zero, there just isn't anything of significance. Now then, the case of public doses from tritium, even the most highly exposed that I can see from tritium is less than 20 µSv, usually as you

move away, doses are down into a fraction of a µSv. So it is very unlikely for one to be able to see an effect from tritium, and to illustrate that I'll show an extract from a natural radiation map of Canada, this is just terrestrial radiation from uranium, thorium and potassium-40. It's actually the area quite near to Chalk River, it's also the area of Renfrew County in Canada, and of course since we are in Renfrew County in Scotland, so I thought it would be quite interesting to show it as well. You can see the contours here is about 100 km^2. The contours here range from about 120 µSv/y, and this is just from terrestrial radiation. You move down street it could change by tens of µSv y^{-1} that you are getting from terrestrial radiation. So the prospect of seeing this signal from something which is giving you only an extra µSv or a fraction of a µSv, is I think quite clearly very remote.

For occupational exposures to tritium, situation may be a bit better. We know exposures have occurred in many facilities. Tritium doses in the studies that have been made being included in some of them, but they generally have not been separated from other exposures. But nevertheless it should be possible to carry out these sort of studies. Unfortunately, tritium by enlarge makes a relatively small contribution to the lifetime dose of workers. It is likely in this sort of study to have a low statistical power. And for some of you realise that in the old days we actually used to keep records on card and on paper. They kind of might be a little unreliable now even if you can find them.

Nevertheless, Little and Wakeford did show some possible data that could be used from the UK facilities, but there really aren't very much, the number of workers are about 5000 workers, collective dose of about 10 person Sv. And even if you take ICRP's nominal risk coefficient, you are not really going to see or expected to see any extra cases. I have added in what we seem to have on tritium doses in the National Dose Registry in Canada, which certainly adds substantially to the doses, takes the total number of workers up to 22,000, 164 person-Sv of collective dose, and maybe a few nominal excess cases. But in such a large population, it is going to be tough to see anything of significance. I would like to note, around the world there are many other cohorts. In South Korea, Romania, India, Argentina, U.S., France, Russia, and China, where there have been probably substantial tritium exposures. And it would be great if any of you here from those countries could persuade the authorities to get that data into some international corporative venture so we could try to put it all together to see if such an epidemiological study might produce.

My final chapter is effluent management. The issue here is, "How do you assess the radiological importance of widely dispersed tritium?" This is sort of getting into

philosophy almost. Way back in the 1970s ICRP suggested that collective dose was an appropriate measure of detriment in radiation. ICRP *Publication 26* suggested that optimisation of protection could be through a cost-benefit analysis. The idea here is that, as shown here: collective dose against the cost of protective measurements. You add protection with increasing cost, and you find the optimum when the marginal cost-effectiveness reaches some chosen value of dollars per person-Sv of collective dose. This is standard cost-benefit analysis. It actually works in occupational settings, but the question raised is that, "Is it applicable to tritium and other globally dispersed radionuclides?", and this is a question asked by the Nuclear Energy Agency (NEA). This is where I have had the privilege of working with Lindell and Benson for the first time to try and answer this question. It came quite clear that the collective dose was made up largely of very small individual doses. So do you really give them the same weight as the large doses? Can you cut them off? Or logically "No", if you follow the linear non-threshold (LNT) model for radiation risk. And people have quite strong opinions on this as you know, and I was rather amused to see a quotation that is from Lindell, Lawrence and Taylor had in its large text on ICRP and NCRP, where Lindell says, "Not adding in small doses had the same misleading character as the belief of Zenon… that Achilles would never beat the turtle". Now since then, the approach to optimisation has been broaden and we are aware that we now talk about matrices, you look at the collective dose in various individual dose sectors, and you make some valued judgement there. But the logic to my mind, LNT still applies, it is still very difficult to say ignore it.

So we have to come back to what Box and Draper suggested and often quoted, "All models are wrong, but some are useful". Certainly LNT is really useful. Dosimetric concepts and quantities depend on the LNT model: additivity of doses, incremental risk being proportional to incremental dose, concept of effective dose. Many other aspects of radiological protection depend on LNT, so it's very useful. The good microdosimetric arguments for initial damage to tissue being proportional to dose at very low doses; and Beninson in the Sievert Lecture I recall from the 9th IRPA Congress, made a very cogent argument along these lines. The trouble is, the discussion of this often has a question ill posed, it's in the form, of what's often called the "sucker's choice", as a response LNT "Yes" or "No". The truth as a matter is much more complicated than that. The probability of radiation carcinogenesis in an individual can well follow a LNT relationship with the magnitude of any single acute small radiation dose. But what we observe in a population is the net of any such carcinogenic events from any such single doses on individuals and any other positive or negative effects on health – Gentner and I was making this point a decade or so ago. Feinendegen and his colleagues have been making the point for quite a while, and most recently in 2011.

It's really, we have an LNT component, but it's actually quite well quantified, but we know all effects – both the LNT ones and all the other effects – are very uncertain at low doses. Even back in 1972, Alvin Weinberg who was head of Oak Ridge at the time in the States, put it in the round of trans-science. He said that, "there's no practical basis for estimating the statistical chances and consequences of the occurrence of these effects for any individual irradiation although we know they occur". And I think that's right, expect I would put a caveat on it that we have no way of knowing a priori what an individual's radiation history will have been at the time of any exposure. So we really don't know how susceptible one is to radiation or what sort of adaption any particular incremental radiation dose may produce.

But we can say, in a population or ask in a population, at what dose and dose rate combinations do the risks of radiogenic cancer start to outweigh the contribution of any stimulatory or adaptive effects to overall health outcome? We can't answer that. I think it's a challenge to experimentalist who insist that LNT isn't the correct model. The challenge is that we need quantitative insights applicable to protection, so we can add in the second component to the model that we use in radiological protection.

There's some implications on this that we can still base prospective radiation protection of individuals on LNT because we have absolutely no idea what these other effects may be or even if they – in that particular instance – will occur. But it does give us a logical justification for cutting off collective dose at low average individual doses, because on average we would expect adaption or whatever other positive effects it might be on average may be having an effect. We don't know what value that cut off should be. Another implication which I find interesting is that the RBE for these different phenomena may be quite different; and so if one starts to use cancer-prone animals – this is often done for radiation experiments – they may actually not be very good models for radiation studies. One may find that the adaptation is much greater in such cancer-prone models than it would be in normal.

Let me close the chapters and go to a summary. What I think I have given the impression is that radiological protection encompasses a very challenging range and a variety of scientific disciplines. It is an exciting field to be in. Given that – this perhaps reflects my own biases – you need a solid grounding in basic physics, chemistry, biology and mathematics, and particularly in statistics. Again also – again I am an experimentalist – you should be prepared to measure, not just model, and be skeptical of models.

Specifically, for tritium, by enlarge the radiological characteristics are sufficiently well understood for most practical health physics purposes, although I have pointed out where some improvements could be made. Many monitors are now, but there could be better discrimination against radiation backgrounds. Dosimetry models could be improved with experimental data on OBT ingestion, on particle inhalation, on intake from surfaces and corresponding interpretation of bioassay results. It would be very useful to have a definitive measurement of RBE for mammalian carcinogenesis for tritium, but I would argue that keeping radiation weighting factor of one is sensible for protection purposes. Continuing intercomparison and validation of models for dispersion in the environment are essential. Certainly, terrestrial and aquatic food chain studies/experimental studies would be very helpful, and in fact are needed for HTO and OBT. I made the point that no effects on health from tritium have been discernable in epidemiological studies. But I have made the point that an international study on the health of workers in many countries does need to be undertaken even though the expected statistical power may be quite low. And finally, appropriate consideration of the small radiation doses to individuals from effluents depends on rethinking LNT. We need to recognize that the LNT carcinogenic response is modulated by other effects, which need to be quantified.

Thank you once again, that's it.

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