IAEA EMRAS, Tritium and C-14 Working Group

EMRAS: Modelling the Transfer of Tritium and C-14 to Biota and Man Notes of the 3rd Working Group Meeting, Baden-Baden, Germany (13-17 September 2004)

EMRAS, Tritium and C-14 Working Group, Meeting Report 3

Notes of the IAEA EMRAS Tritium and C-14 Working Group Meeting, Baden-Baden, Germany (13-17 September 2004)

History:

The third meeting of the IAEA EMRAS Tritium and C-14 Working Group was held in Baden-Baden, Germany, as part of the 7th International Conference on Tritium Science and Technology (Tritium 2004). The meeting was hosted by Forschungzentrum, Karlsruhe.

These Meeting Notes have been prepared by A. Venter (Technical Secretariat), P. Davis (Working Group Leader) and M. Balonov/T. Cabianca (Scientific Secretariat). In addition, the following people who attended the meeting contributed to the discussions and decisions documented in these Meeting Notes:

Name	Organisation	Country
J Aign	Munich Technical University	Germany
M Atarashi-Andoh	JAERI	Japan
F Baumgartner	Munich Technical University	Germany
D Cutts	Food Standards Agency	UK
D Galeriu	IFIN	Romania
V Golubeva	VNIIEF	Russia
P Guetat	CEA	France
Y Inoue	NIRS	Japan
H Kakiuchi	Institute for Environmental Sciences	Japan
J Kozar Lagar	Jozef Stefan Institute	Slovenia
S Mavrin	VNIIEF	Russia
N Momoshima	Kumamoto University	Japan
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W Raskob	Forschungzentrum Karlsruhe	Germany
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Introduction

The third meeting of the EMRAS Tritium and C-14 Working Group was held in Baden-Baden, Germany, on 13-17 September 2004. It was hosted by Forschungzentrum, Karlsruhe, as part of the 7th International Conference on Tritium Science and Technology (Tritium 2004). The objectives of the meeting were the following:

- discuss the draft reports for the Perch Lake and Soy Bean Scenarios;
- present and discuss preliminary results for the Pickering Scenario, and for the scenarios involving hypothetical short-term tritium releases;
- finalize the Pine Tree Scenario;
- present qualitative results from the experiments on fresh water mussels undertaken at Chalk River Laboratories and decide whether the data can be used as the basis for a scenario;
- continue discussion of data sets involving tritium dynamics in large animals and fish;
- continue discussion of C-14 data sets and decide on C-14 scenarios;
- plan future work activities.

Participants were welcomed to the meeting by the Working Group Leader, Phil Davis. Participants introduced themselves and briefly described their background and interest in the working group.

Draft report on the Perch Lake Scenario

Phil Davis gave a brief summary of the Perch Lake Scenario, and then tabled the draft report, which presently contains introductory material plus an analysis of the results for algae and bullheads. Sections on worts, cattails, clams, pike and sediments are still to be written. The following points emerged from the discussion:

- Most modellers assumed HTO concentrations in algae were equal to local water concentrations and obtained results in good agreement with the observations.
- Predicted OBT concentrations in algae showed greater variability due to different choices for the isotopic discrimination factor.
- The OBT/HTO ratio in algae is different for UK and Canadian data. The reason for this is not clear.
- The observations show that HTO concentrations in bullheads are the same as the average concentration in all bottom waters, since bullheads are benthic fish (bottom feeders). On the whole, predicted HTO concentrations in bullheads were in good agreement with the observations; the few cases of large differences were due to incorrect assumptions regarding the degree to which the fish draw tritium from the sediments.
- Most modellers assumed OBT concentrations in bullheads were proportional to the HTO concentrations and did not consider what the fish eat, or OBT formation in the

fish. None of the modelers did any sort of time-averaging when calculating OBT concentrations, even though the metabolic turnover for OBT is slower than for HTO. The predictions showed considerable scatter among models and 4 of the 8 results did not agree with the observation even when uncertainties were taken into account.

- Uncertainties: The magnitudes of the uncertainties in the predictions varied greatly among models and were very high for some models. The uncertainty results for all endpoints will be investigated in more detail in the future.
- The observed OBT concentration in fish was larger than the observed OBT concentration in plants. This could be because OBT formation in fish is due to metabolic processes, though such formation is expected to be low. Alternatively, OBT residence times may be different in plants than in fish. The tritium concentrations in the lake show a slow decrease over time and the high OBT concentration in fish (compared to that in plants) may be a memory effect.
- The observed OBT concentrations in sediments, which are made up primarily of decaying plant material, were only about half the concentrations in fish or plants. This is probably because OBT is broken down to HTO when the plants decay/decompose.
- Some modelers predicted different tritium concentrations in different parts of the fish, although the data show clearly that this is not the case.

There will be an appendix with a description of each model in the final TECDOC.

Draft report on the Soy Bean Scenario

The draft soy bean report was presented by Phil Davis in the absence of the scenario leader, Hansoo Lee. The current version of the report includes the scenario description, the observed HTO and OBT concentrations, a graphical comparison of predictions and observations and a brief discussion of some of the results. Discussion in Baden-Baden centred on a number of questions that Hansoo put forward prior to the meeting.

Participants questioned whether the initial observed ratio of the HTO concentration in plants to the HTO concentration in air moisture was correct/sensible, since it was quite low. This ratio depends on temperature and it was concluded that the ratio could be reasonable given the observed high temperature in the chamber during exposure. However, the models tend to overpredict the observed value.

Most models underpredicted the HTO concentration in plants towards the end of the simulation period. The reason for this is not entirely clear. The concentrations may remain high due to the breakdown of OBT. Alternatively, some HTO taken up by leaves may be immediately sequestered in the roots and stems and recycled back to leaves during the fruiting period. There is some storage in different parts of the crop but this is difficult to model. The differences in the observed time-dependent behaviour of the HTO concentrations in experiments SB1 and SB4 are likely due to the different growth stages of the plants during exposure and the different climate conditions experienced by the plants after exposure.

Tritium concentrations in the plants are expected to depend strongly on the growth rate in the non-linear stage of growth, but less so in the linear phase. There was some concern that the growth models used in the codes, which reflect an average plant, do not describe very well the individual plants used in the study, which varied quite a bit from one to another.

The predictions vary greatly from model to model. This needs to be resolved. The scenario leader needs more information from modellers on their conceptual approaches and model parameter values.

First round of calculations for the Pickering scenario

The scenario leader, Phil Davis, gave a short summary of the Pickering Scenario. Results were received from 6 participants, and those present at the meeting gave short overviews of their models.

Results were presented for soil, animal feed, garden vegetables and fruit, chicken products and cow products. For a given model, predicted/observed ratios for all samples in a given category were averaged over all sites and over both sampling periods.

Soil: The predictions were within 20% of the observations for most models.

Forage crops: The predictions were good for DF8 in July but the models overpredicted at other sites and times by a factor of more than 2 on average. The overpredictions were largest for F27, where there is some concern that the observed air concentrations are too high. When the results for F27 were left out of the average, the agreement between predictions and observations was better, but the models still overestimated the concentrations in forage crops by about 75%.

Fruit and vegetables: All modellers overpredicted the observed concentrations by a factor of more than 3 on average. The overprediction was greatest for root crops, followed by fruit, non-leafy vegetables and leafy vegetables. Since fruit and vegetables were sampled only at F27, the differences between predictions and observations could be due in part to uncertainties in the air concentrations.

Animal products: The P/O ratios for chicken and eggs (sampled at F27 only) ranged from 0.5 to 4.5; and those for milk and beef from 0.5 to 3. The ratios were essentially the same for chickens and eggs and for beef and milk. For beef and milk, the modelers had to take into account the fact that the cows ate crops grown in the previous year.

Because some submitted results were clearly in error, and because other modelers have expressed an interest in participating in this scenario, the observed data will be released only at the end November 2004. The scenario will remain open and new or revised results can be submitted until this time.

Scenarios for hypothetical short-term releases

The purpose of modelling hypothetical scenarios is to provide guidance to decision makers in the event of an acute tritium release to the atmosphere. The first 24 hours would be the most crucial in terms of HTO; OBT would be of importance at later times when foods are harvested. The assumption for the first round of calculations is that 10 g of tritium (as either HT or HTO) is released over a period of 1 hr. The calculation period is 1 year. Seven participants submitted results.

The scenarios aim to answer questions such as the following:

- Based on total dose without intervention, are protective measures required at any downwind distance within 30 km of the release point?
- Do doses to both children and adults need to be considered?
- What vegetables should be banned from public consumption?
- How long must agricultural activities be suspended before crops can once more be planted in contaminated soil?
- How long do cows have to be removed from pasture?
- What should a post-release monitoring plan consist of?
- How should contaminated products be disposed of?
- What is an appropriate reference activity[1] at which the international trade of contaminated products would be restricted?

Limits on trade are normally based on concentrations in foods and are deduced from the relevant dose limits. (An IAEA Safety Guide on this issue is being produced. Further information can be obtained from Mikhail Balonov.) This is a meaningful process for radionuclides that move slowly in the environment, but is not appropriate for tritium, for which concentrations vary rapidly over time. Tiberio Cabianca suggested that the EMRAS Tritium WG is not the appropriate forum for discussing trade limits or intervention levels.

The initial results of the first round of calculations were presented by the scenario leader, Philippe Guetat. Results were normalised to air concentrations, deposition to crops, and deposition to soil, as appropriate, to reduce the impact of errors in one part of the calculation on subsequent parts.

<u>Case 1</u> (HTO release in unstable conditions)
The air concentrations predicted by the various models varied by a factor of 10 at a downwind distance of 300 m and a factor of 100 at 30 km. This is believed be due primarily to the choice of stability class, with differences in roughness length and

[1] Tritium reference level for international trade in foods is 10 000 Bq/kg (to be approved).

dispersion parameters also contributing. All of these variables need to be defined more precisely.

Case 2 (HTO release in neutral conditions during rain)
The results agreed to within a factor 2-5.

Case 3 (HTO release in stable conditions)
Air concentrations at 1 km varied by 2-3 orders of magnitude, which is again likely due to the choice of stability class, roughness length and dispersion parameters.

The distribution of ingestion dose varied greatly between models. In the German and French models, the largest contributor to ingestion dose was cereal; in the Japanese and Korean models, it was garden vegetables; and in the Canadian model it was meat and milk. It was noted that Japanese parameter values were different from European values, even though consumption values were specified in the scenario. There was little difference between modelled adult and child doses. Given the large variation in the predictions, it was decided to delay a detailed discussion of the results until a second round of calculations could be carried out with a more tightly defined scenario.

One of the aims of the exercise is to identify those processes that need to be included in the models. It was decided that an interaction matrix (a matrix classifying the processes with respect to importance and documenting the way they are treated (or not treated) in the various models) could be helpful.

Results for HT releases were not discussed, but HT inventories are too small to be significant in terms of dose. The revised scenario description will address the amount of tritium assumed to be released.

The revised scenario incorporating the additional information will be distributed at the end of October for the next round of calculations. Crops can be grouped into broad categories (root vegetables, green vegetables, etc) if desired. Infant doses will be requested for one case only, to be decided by the scenario leader. The Technical Secretariat will provide information on the processing factor for cheese. Results should be returned in the format specified no later than 20 January 2005.

Pine tree Scenario

Yoshi Inoue gave an overview of the Pine Tree scenario, which has changed since the last meeting to incorporate groundwater information. Release rates are monitored at two facilities, JAERI (HTO only) and JNC (both HTO and HT) and endpoints are calculated primarily at monitoring point MS2. A revised scenario that takes into account comments made at the meeting will be issued during the first week of October 2004. Modellers should return results by 15 February 2005.

Additional tritium test scenarios

Experiments with animals

Birgit Wierczinski (Technical University, Munich) is liaising with the **Russian** Institute of Agricultural **Radiology and** Ecology, Obninsk, –with the aim to undertake tritium experiments with animals under the ISTC programme. Birgit gave an update on the contact with Obninsk, where it is possible to do experiments (inhalation and ingestion) with chickens, sheep and cows (see attached document). A major factor in the discussions was the ethics of performing such experiments, including the need to keep the number of animals involved to a minimum and the need to design the experiments to minimize the number of animals sacrificed.

Chickens:

The meeting felt that transfer via inhalation is too low to justify killing animals. The tritium concentration resulting from this pathway can be evaluated from analysing urine and blood (weighing chickens with every analysis), keeping in mind that OBT in blood is a conservative estimate of OBT in meat. It would also be important to determine the water balance of the chickens (i.e. respiration rate, air moisture and temperature). It was finally decided that the inhalation pathway for chickens should not be included in the experiments.

Further discussion centred on the ingestion pathway, and whether special grain would have to be grown, how long the chickens should eat the grain (instantaneous vs. continuous ingestion) and how the chickens should be fed (natural grazing vs. intravenous). To minimize stress conditions the chickens should be allowed to graze naturally. Chickens would ingest both HTO and OBT (in the grain). Local radiation protection procedures should be considered when producing the contaminated grain. The background tritium concentration in air should also be determined, as any tritium in the air will contribute to tritium in the grain and the animals, even if very little. The tritium concentration in the grain should be uniform and sufficiently high to allow concentrations to be measured with little uncertainty. The number of chickens that have to be sacrificed can be reduced by emphasizing eggs in the study.

Sheep:

The meeting decided that sheep do not need to be sacrificed to determine the contribution of the inhalation pathway. The inhalation rate of sheep should be measured, and the concentration in meat can be approximated by analysis of blood and urine.

To determine the contribution of the ingestion pathway, and to follow the build-up of tritium in the body, sheep probably need to be fed continuously for 2-3 days. Once the feed has been stopped, tritium decay can be followed. There was considerable discussion on how the feed should be administered and whether instantaneous or continuous feeding would be easier from a practical point of view. HTO and OBT (i.e. water and food) need to be administered in separate experiments.

It was suggested that pigs should be used instead of sheep, as the metabolism of pigs is nearest that of humans. However, the pig diet is highly variable and it might be difficult to tritiate the required crops.

Cows:

The discussion and conclusions for cows was much the same as for chickens and sheep. One issue unique to cows is whether they should be fed fresh grass containing both OBT and HTO, or cut grass that has been stored and therefore contains mainly OBT.

General:

There is a need to obtain more details about the experimental setup, sample analysis, and the detection levels and uncertainties in the analytical techniques. Such an experiment would require a large amount of money and therefore must be designed properly. The meeting decided that there is value in using an animal tritium model to assist in designing the experiment. For instance, a model could be helpful in evaluating how much tritium needs to be administered in order to ensure that the tritium concentration in the final animal sample is such that it is above the detection limit. A model can also be useful in deciding the frequency of sample taking, and in optimising the number of samples that need to be taken (i.e. the number of animals that need to be sacrificed). Dan Galeriu volunteered to use his model to provide this type of information.

It is hoped that ISTC funding will be obtained within one year. It is possible that the experiment will not be completed within the EMRAS lifetime.

Experiments with crops

Valentina Golubeva gave a presentation on the tritium exposure experiments being conducted at VNIIEF (see attached). The purpose of the experiments is to study OBT build-up in crops. Tomatoes, leaf lettuce, potatoes and spring wheat grown under field conditions were exposed to HTO in a greenhouse for 2 hours, and then returned to the field until harvest. Each crop was grown in three areas, one for producing plants for exposure, a second for background measurements and the third for growth rate determination. The plants were sampled and the OBT and TFWT concentrations were measured following the methods recommended by the IAEA in 1961.

Comments are invited on the experimental and analytical procedures and should be sent directly to Alexey Golubev (avg@dc.vniief.ru).

Experiments involving aquatic organisms:

Phil Davis presented details of experiments with freshwater mussels exposed to tritium at Chalk River during the summer of 2004. The purpose of the experiments was to provide data for testing models that predict the evolution of HTO and OBT in aquatic organisms exposed to an abrupt change in ambient tritium concentrations. Mussels were obtained from the Ottawa River upstream from Chalk River, transported in buckets to Perch Lake (travel time about 0.5 hr) and transplanted into cages in the lake. 64 mussels were transplanted into each of 4 cages, two of which had bare bottoms and 2 of which had 10 cm of lake sediment on their bottoms. From visual inspection, the mussels were functioning normally soon after transplantation. All mussels were 9-11 cm in diameter, but their age was unknown. The mussels were sampled every hour for the first two hours after transplantation and then at increasing times up to 50 days. HTO and OBT concentrations and the mass of soft tissue were measured in each sample. Water samples were taken every day and sediment samples were taken at the same time the mussels were collected. There is a possibility to follow tritium decay in the mussels by moving 'old' mussels from Perch Lake to the Ottawa River next year. A draft scenario description should be available at the spring meeting.

It would be useful to have additional data sets on fish but none is known. Experiments with larger fish are difficult and costly. The possibility of a study involving a commercial fish farm cage in Cardiff Bay was discussed but in the absence of the representative from GE Healthcare no commitment could be made.

Definition of OBT

Yves Belot and Phil Davis suggested the following definition of OBT for consideration by the WG:

Conceptually, OBT is carbon-bound tritium that is originally formed in living systems through natural environmental or biological processes from HTO (or HT via HTO). In practice, OBT concentrations are best determined experimentally as the activity of non-exchangeable tritium in the combustion water of the sample in question. The experimental value must be corrected if the specific activity of tritium in carbon-bound hydrogen is required. Other types of organic tritium (e.g., tritiated methane, tritiated pump oil or radiochemicals) should be called tritiated organics, which can be in any chemical or physical form.

This definition was accompanied by several notes that provided further information and clarification.

It was suggested that the definition would be more accurate if the words "conceptually" and "in practice" were deleted from the first and second sentences respectively. This was accepted.

Most of the further discussion of the definition revolved around the concept of buried hydrogen. The experimental work of Franz Baumgartner suggests that a significant amount of the tritium that remains in the dry matter after washing is not carbon-bound, but instead is attached to large molecules that do not participate in the exchange process. Such molecules are quickly broken down once taken into the body and the associated tritium subsequently acts as HTO rather than OBT. Buried tritium must be considered part of OBT if the analytical techniques presently used to measure OBT are to be preserved. However, it is not clear if the dose coefficients currently in use are consistent with such a definition. They may be since the coefficients are based largely on empirical loss rates observed after ingestion of OBT, which includes buried tritium. Phil Davis will look into this question with dosimetry experts at Chalk River.

C-14 datasets

The following datasets were identified in previous WG meetings:

- EDF data: These involve a continuous release of C-14 to the atmosphere. Endpoints would be C-14 concentrations in ivy leaves and grass. However, few of the plant concentrations were obtained at the locations where air concentrations were measured, and the availability of meteorological data is unknown.
- Imperial College data: These involve plant exposures in chambers similar to the soybean scenario. Endpoints would be C-14 concentrations in cabbage, potatoes and beans. The data appear to be of high quality and are readily available.
- Pickering data: C-14 concentrations are available in all of the samples collected at Pickering for the Pickering tritium scenario. However, concentrations were only slightly above background and it would be difficult to use them to draw definitive conclusions about model performance.
- Wetland data: Data are available in groundwater, surface water, soil (decomposing organic matter), air, vegetation and wild animals (small mammals, amphibians, snakes and insects) at a wetland site on Chalk River property that is contaminated by discharge of C-14 from a waste management area.
- JNC data: These involve a continuous release of C-14 to the atmosphere. The endpoint would be C-14 concentrations in rice. All the required information, including meteorological data, is available and of high quality.
- The Centre for Ecology and Hydrology (UK) has carried out experiments with C-14 on sheep. Part of the data has been published.

Data on C-14 in tree rings in years before and after the Chernobyl accident are also available. However, release rates from Chernobyl were not well defined. The release continued over 10 days and the meteorological conditions were complex. This scenario is not really suitable for this group as it would involve too much dispersion modelling.

Following discussion and expressions of interest by the participants, it was decided to first model a scenario based on the JNC data, followed by a scenario based on the Imperial College Data.

Future Work Programme

ITEM	ACTION	PERSON	DATE
Hypothetical scenario	Revise scenario Submit results from second round of calculations Present results	P Guetat Modellers P Guetat	30 October 20 Jan 2005 Spring meeting
Perch Lake Scenario	Complete draft report Comments Final report	P Davis Participants P Davis	30 Jan 2005 30 March 2005 Spring meeting
Soy bean scenario	Final report Comments Final report	H. Lee Participants H. Lee	30 Jan 2005 30 March 2005 Spring meeting
C-14 Rice scenario (tentative)	Draft scenario	J Koarashi	Spring Meeting
Pickering Scenario	Submit revised/new results Submit model descriptions Distribute observed data Draft report	Modellers Modellers P Davis P Davis	30 Nov 2004 30 Nov 2004 30 Nov 2004 15 April 2005
Pine tree scenario	Revised scenario Results Present results	Y Inoue Modellers Y Inoue	7 Oct 2004 15 Feb 2005 Spring meeting
Definition of OBT	Revise definition	P Davis	Spring Meeting
Animal experiments	Liaise with Obninsk	B Wierczinski	Ongoing
Mussel scenario	Complete discussion of draft scenario Finalize scenario	T Yankovich/AECL T Yankovich	Spring meeting Plenary meeting 2005

Next Meeting

The next meeting is provisionally scheduled for Cardiff (UK) in mid to late April 2005.

Further Information

Information on the activities within EMRAS generally and on the Tritium and C-14 WG in particular (including the scenarios being used for model testing), can be obtained from the following people, respectively:

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ANNEX A: List of Participants at the 3rd EMRAS Tritium and C-14 Working Group Meeting, Baden-Baden, Germany (13-17 September 2004).

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