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Modelling the transfer of ^{3}H and ^{14}C into the environment – lessons learnt from IAEA's EMRAS project

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Abstract. Future development of nuclear energy in the frame of climate change and sustainable development needs an increased safety and consequently, robust models of environmental transfer of radionuclides. Tritium and Carbon are life elements and must be treated separately from trace elements. The IAEA promoted EMRAS (Environmental Modelling for Radiation Safety) project in order to decrease uncertainties in the predictive capability of environmental models, including the cases of aquatic and biota. To understand the processes and models reliability, nine scenarios have been developed. The Working Group contributed to the Revision of "Handbook of parameter values for the prediction of radionuclide transfer in temperate environments" TRS 364, as well. The main task of this paper is to propose ways for models' predictive power improvements, based on lessons learnt from EMRAS' exercises.

1. INTRODUCTION

EMRAS continues some work of previous international programs in the field of radioecological modelling. The activities of EMRAS were focused on areas where uncertainties remain in the predictive capability of environmental models. Working Group on Modelling of tritium and carbon-14 transfer to biota and man focused on improving models of OBT (organically bound tritium) formation and translocation in plants, animals and fish. The Working Group necessarily had to consider HTO (tritiated water) as well, since an understanding of environmental HTO is needed before OBT can be modelled. Carbon-14 was also on the agenda since the dynamics of carbon and OBT are very similar. In the activities of the T&C Working Group, routine and accidental releases using nine blind tests or modelling inter-comparisons have been considered (Table 1), and we participated to all of them.

Table 1. The scenarios developed in the frame of working group.

Scenario	Type	Scenario Leaders	Participants
Soybean	H-3, Accidental, soybean plant	KAERI Korea	Canada, France, Germany, Japan, Korea, Romania, Russia, UK, USA
Perch Lake	H-3 Routine, aquatic	AECL Canada	Russia, France, Germany, Romania, Japan, UK, Lithuania
Pickering	H-3 Routine, terrestrial	AECL Canada	Germany, USA, Lithuania, UK, Romania, Japan, France
Pine tree	H-3, routine, tree, groundwater	NIRS Japan	Japan, USA, Romania, France
Rice	C-14, routine, rice	JAEA Japan	Canada, France, Romania, Japan
Hypothetical	H-3, accidental, terrestrial	CEA France	Canada, Romania, Korea, Japan, France, Germany, India
Mussel	H-3, accidental, aquatic - mussel	AECL Canada	Germany, France, Japan, Romania
C-14 potato	C-14, accidental, potato plant	NIPNE Romania	France, UK, Romania, Japan
Pig	H-3 (OBT) - complex - pig	NIPNE Romania	Japan, Romania, France, Canada, UK

An important contribution of the Working Group is participation to the Revision of "Handbook of parameter values for the prediction of radionuclide transfer in temperate environments" TRS 364, which includes routine release where the present assessment reliability is acceptable.

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2. THE DISTINCTION BETWEEN ROUTINE AND ACCIDENTAL CASES FROM THE POINT OF VIEW OF MODELLING APPROACH

In classical routine operation of nuclear installations, it is considered constant releases of radionuclides, constant (average) dispersion from source to receptor, and equilibrium conditions in all compartments of an ecosystem. For tritium and carbon, specific activity approach is used, in order to deduce transfer factors or concentration ratios in food items and surrounding environment. In practice, the release of radionuclides is not constant, meteorological conditions are not constant, and subsequently, concentrations in food products are not in equilibrium. The modelling approach must include the quasi-equilibrium conditions and the distinction between processes, which are strongly influenced by change of environmental conditions and other processes, which temporally integrate the variability of environmental factors.

For an accidental emission, the models must carefully take into account the influence of environmental factors on the transfer processes of radionuclides. Tritium and carbon-14 are life elements, and for these radionuclides it is necessary to take into account the adaptive response of the organisms for environmental constraints. Hydrogen has a relatively complicated behaviour depending on its initial chemical form, its transformation from one form to another and instantaneous climatic and soil conditions. Many processes are in common with ^{14}C, a radionuclide less difficult to model because the influence of water cycle is better understood. It is recommended that tritium modelling to start with a good understanding and modelling of ^{14}C processes. For more details see the dedicated chapter.

EMRAS considered many "routine scenarios" and variability of source term and meteorological condition were studied for the first time in multi-annual assessment. The ^{14}C in rice scenario and the pine scenario referred to coastal site and NO model has considered the influence of sea breeze on the atmospheric dispersion. This demonstrates the insufficient flexibility of our present models and needs for further improvements. Yearly variation in the climatic conditions influences plant growth and combined with variation in the source term, can explain long term variability in crop concentration at harvest. As it is shown in the rice scenario, ignoring the real dynamic of plant growth and the detailed ^{14}C uptake can induce a misprediction up to a factor 10 (see Figure 1 for 1992).

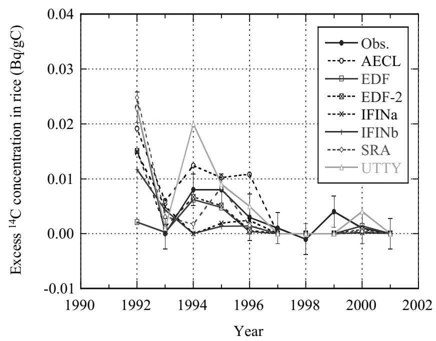

Figure 1. ^{14}C concentrations in rice at sampling site R-1 from 1992 to 2001.

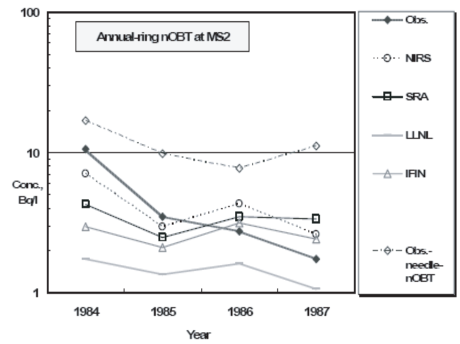

Figure 2. Observed and predicted yearly variations of OBT concentrations in annual-rings at MS.

Concerning the pine tree scenario, where tritium was released as HTO, it may see also that the mispredictions can be partially explained by the conversion processes from HTO to OBT. Plant physiology such as the difference in a photosynthesis rate between daytime and night time and between seasons, and translocation of OBT are supposed to be quite important to predict the OBT concentration in plants. An interesting fact to note is that the OBT concentration in annual tree rings is lower by a factor 2 than OBT concentration in pine needles (see Figure 2) and the causes must be further explained. As conclusions concerning pine tree scenario, we learned a few important features by prediction-observation comparison exercise: i) the air concentration, which is primarily important for prediction of the rest of sample species, is affected by local meteorology such as fumigation effect when discharge sources are located along the sea coast, ii) the TFWT (tissue free water tritium) and OBT concentrations are affected by daytime meteorology, and iii) the plant OBT concentration is affected by plant physiology of OBT production, its translocation and associated hydrogen isotope effect. All of these features should be further studied in future for confirmation.

It merits to be pointed out that it is necessary to check internal consistency of model prediction in respect with basic knowledge on processes, as well as the consistency of the predicted to observed ratio. In above scenarios as well as in the Pickering one, the main conclusion was the peculiar importance of a good modelling/monitoring of air and precipitation concentrations. In Pickering scenario it was shown that the daylight air concentration is 2–3 times lower than 24 hours concentration average. OBT is produced more intensively in the light and all models' predictions are improved if it is used only daylight concentration. This cannot be generalised because we have a poor knowledge of OBT formation in night time in many plants. This must have two major influences on further work: a) for operational purposes, after the screening of potential sites for "reference person" [1], on site air monitoring must be mandatory considered at potential sites; b) for licensing purposes, improved models for atmospheric dispersion and washout must be considered based on up to date approaches and abandoning the Pasquil-Gifford scheme.

For routine scenarios, in the last 10 years, at international exercises (BIOMOVS, BIOMASS) using monitoring data, we succeeded to have a harmonised view and the models' predictions are reliable at factors 2–3. Further improvements must concentrate on transfer following soil contamination and some gaps in the data must be covered concerning the relative importance of air and soil contributions to the concentrations in fruits and root crops. In case of ^{14}C in soil, more data are needed for the canopy dilution factors as functions of canopy characteristics, atmospheric conditions, and size of contaminated soil sources [2].

An intermediate case between routine and accidental situation, there was mussel scenario with two phases: uptake and depuration. In both cases, an abrupt change of environmental tritium concentration has been considered (constant low to constant high concentration; Heaviside function). The mussel specie used in the study was *Elliptio complanata*. In the uptake case, all models under-predicted the OBT concentration in the mussels one hour after transplantation, but over-predicted the rate of OBT formation over the next 24 hours. In addition, the subsequent dynamics were not well modelled, although all of the participants predicted OBT concentrations that were within a factor of 3 of the observation at the end of the study period. All models over-predicted the OBT levels at the final 88-day experiment time point, likely due to the loss of OBT by female mussels during egg production and release. In respect of misprediction in early time, two explanations are possible: i) as for other animals, the metabolic biokinetic rates have a slow structural and a fast functional component. If the fast component predominates, OBT concentration is higher in early time than for the single average metabolic rate [3]; ii) the mussels have been measured with the stomach content included. Immediately after the transplantation into the higher level tritium environment, activity in stomach increases abruptly, but much slower in mussels' tissues.

For depuration case, the mussels have been exposed to sudden and significant decreases in tritium concentrations into the water, after long equilibration with heavy contaminated water. Mussel HTO concentration equilibrates with the low water concentration in short period but mussels OBT concentrations did not reach steady state over the 117-day period of this study. If stomach content is the only explanation in the uptake phase, we expect a sudden decrease in the first days for the depuration phase, but it is not the case. Complex dynamics of metabolism must be taken into account. Models with a single metabolic biokinetic rate or with stomach content are unable to reproduce the experimental data (see Figure 3). NIRS model with two biokinetic rates better succeeds. IFIN model, with a single biokinetic loss rate ($0.022\,d^{-1}$), giving acceptable results in the uptake phase, largely mispredicts the depuration case. Decreasing the loss rate at $0.04\,d^{-1}$, the agreement is good. There is an unnatural discrepancy between uptake and depuration model results and it is clear that a single organic loss rate is not enough. A compromise can be obtained with single loss rate of $0.011\,d^{-1}$ and the stomach content contribution. Complex models are not recommended for radiological assessment due to paucity of input data or excessive cost to obtain. A compromise must be obtained for an operational model but the problem is to have a control on model predictive power (uncertainty included). The EMRAS mussel scenario was very helpful in order to assess and improve the model predictive power.

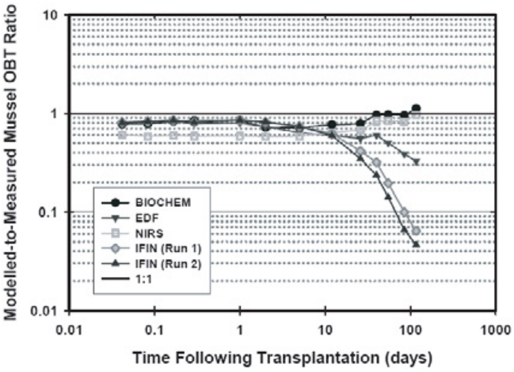

Figure 3. Inter-model comparison of modelled-to-measured OBT concentration in soft tissues of transplanted mussels.

3. NEED OF INTERDISCIPLINARY APPROACHES

The broad range of scenarios as well as the limited time budget available in the EMRAS precludes a deep, detailed analysis of causes of mispredictions or model intercomparison differences. It will be briefly analysed few key points just to demonstrate the need of an interdisciplinary approach in order to decrease model uncertainties.

In the Perch Lake scenario it was observed the importance of spatial average of water concentration for predicting HTO concentration in fish. The correct averaging procedure depends on fish habits and this implies more specific knowledge. Note also that none of the models succeed to predict OBT concentration in sediment due to absence of appropriate scientist in the modelling team.

The Pickering scenario provided a good test of models that predict tritium concentrations in various compartments of an agricultural ecosystem at steady state. The modellers used air concentrations averaged over different time intervals to drive their models. The scenario leader is an outstanding scientist in atmospheric transport modelling and in tritium transfer, too. Drawbacks of air concentration measurement were recognized and causes of over prediction of OBT were scrutinized (by an average factor of 1.9 at the dairy farms and 3.4 at F27 – a hobby farm). Many potential causes were analysed and some are: what air concentration for OBT prediction must be considered – daily average or daylight; how much and how many models include OBT formation at night. To understand OBT formation in night it is necessary to expand our knowledge data base with biochemical processes in plant (light and dark reactions, C3 and C4 plants, role of substrate produced in the day for the dark reaction when hydrogen is bounded in organic forms). In practice also, it is necessary to balance the higher air concentration in night with lower canopy conductance in order to detect the share of tritium uptake in the night with those previously taken in the day.

In EMRAS there have been also scenarios involving specific "controlled" experiments: soybean, mussel, potato. When models must predict the dynamics of radionuclide in specific experiments, it is mandatory to understand the experimental condition. In soybean scenario all models over predicted

HTO in leaves at 0.2 h after the end of exposure and this can have two explanations, not considered by modellers: a) rapid flushing of air from the experimental box at the end of exposure; b) higher temperature in the experimental box than normal – with a potential decrease in photosynthesis and canopy conductance. At harvest, as it was reported in the soybean scenario, the plant body and the beans are still quite wet (60% water in ponds), a situation revealing incomplete maturity. Few modellers realised this in order to better asses the plant development stage at each fumigation period. In the potato scenario, planting was very late and all development was affected, comparing to normal practices. Many models ignored it.

In both soybean and potato scenarios, predictions were asked for storage organs and plant body (leaves or leaves and stems) at various development stages. Mispredictions were quite often at early or late stage of development. It seems that better understanding of plant growth processes are needed in each modelling team. At start formation of storage organs there is a translocation from labile assimilate in stems (accumulated in time) to storage organs and this gives higher concentration in storage organs than assessed by simple growth. Partition of new assimilate (and associated ^{14}C or OBT) into leaves, stems and storage organs depends on development stage AND on genotype, also. Romanian modellers exercised in soybean and potato scenarios two plants' genotypes – one standard for Central Europe and one more adapted to local condition (a Scottish potato cultivar and a Japanese soybean). The last one gives definitely better results when it was compared to experimental data. It is clear that uncertainty can be decreased when it is considered site specific conditions for environment AND genotype.

Pig scenario is other example for the need of interdisciplinary approach in order to obtain reliable operational models for tritium transfer in animals. Models participated to this scenario have various complexities, using a single organic compartment or more, with various assumptions. The models' results have been compared to experimental data for a sow fed 84 days with OBT. It was shown that models with two or more organic compartments give predictions better than a factor 3 for OBT in pig' organs, but intercomparisons for other intake scenarios indicate larger discrepancies between models. The preliminary analysis indicates the need of a better knowledge of nutrition and metabolism of animals, especially the farm ones, to be able to robustly predict the concentration in meet or milk in various intakes' situations.

4. ADVICES FOR IMPROVING TRITIUM ACCIDENTAL RELEASE MODELS

If present accuracy in predicting concentration in products under routine conditions seems satisfactory and it is agreed on some standardization, the case of accidental release needs more efforts. In order to better handle the uncertainties, it will be useful to develop a harmonized approach, not a standard in *stricto senso*, because processes are widely variable under local conditions. It seems clear that results presented in EMRAS, as well as in previous projects (BIOMOVS, BIOMASS) are scenario specific and difficult to generalize to other processes, sites and some models were developed for specific tasks and cannot be easy generalized. Model performance does not seem to be improved over time and this must be changed now under the revival of nuclear energy. Because a major contribution to overall uncertainty is due to atmospheric dispersion [4], any new model must include more reliable atmospheric transport and dispersion models that take turbulence data and topography into account and also have a good representation for reemission (improved area source). While it is recognised that HT emissions are less harmful than HTO emissions, the process of HT conversion to HTO in soils must be analysed starting from basic science and modelled accordingly with local soil properties. The influence of environmental condition on the transfer of tritium to plants must be included and generic models must separate wet, dry, and hot or cold situations. In all situations the model must have the ability to take environmental conditions into account in a dynamic way. For the transfer of radionuclide in the atmosphere-soil-plant-animal continuum, all sub-models should be based on physical approaches and use knowledge from other disciplines to derive general dependencies based on available data for substances other than tritium. Knowledge from agricultural science must be incorporated as, for example, crop growth modelling based on plant physiological parameters and processes (photosynthesis, partition of newly formed dry matter, genotype influence, evapotranspiration). It is mandatory also, to consider recent progresses in understanding of carbon and hydrogen cycling and use this in ^{14}C or ^{3}H models. It must be noted the recent progress in Crop and Weed Ecology [5] and the link between photosynthesis and canopy resistance. For OBT production at night it must develop an improved model based on a deeper analysis of plant processes and not simple fitting of peculiar experiments. Translocation in fruits and roots must be modelled using knowledge in agricultural research and tested only with experimental data, if they are available. The recent generic model based on energy metabolism for the transfer in mammals must be further developed for operational application, because many experimental data are missing. The predictions for contamination of eggs or broilers must be experimentally checked. The dosimetric applications of the models must include infants and pregnant woman, to be able to reliably assess the dose for public, in the present debate on tritium risk. For cold climate, tritium behaviour in winter, including washout by snow, dry deposition to snow and the fate of tritium in the snow pack must be studied and robust models must be developed. A further reduction of uncertainties must be based on the ability to use site-specific information on land use, local soil properties and predominant crop genotype characteristics, together with realistic assumptions concerning habits of the maximally exposed individual.

5. CONCLUSIONS

We discussed the most important lessons learnt from EMRAS related to ^{3}H and ^{14}C cycling in the environment and their related models and it seems that our understanding and the predictive power of radiological models is still limited by large uncertainties, especially for accidental releases. Further experimental work on OBT production in night time is needed, because it is a key process under debate for both routine and accidental release. Tritium transfer, mostly as OBT in broilers, eggs, and large fish are also desired, because models' uncertainty on this topic is difficult to assess without experimental data. It was established that no process can be ignored *a priori* due to the large variability of environmental and site-specific conditions at the time of the accident. In order to harmonise our understanding at process level, a large interdisciplinary effort must be done and this needs communication between radioecology scientists and other life scientists. Nuclear energy is again considered a viable energy source for the future and this necessitates continuous improvements of radioecological models.

Acknowledgments

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