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Uncertainty of current understanding regarding OBT formation in plants

A. Melintescu*, D. Galeriu

"Horia Hulubei" National Institute for Physics and Nuclear Engineering, Department of Life and Environmental Physics, 30 Reactorului St., P.O. Box MG-6, Bucharest – Magurele, RO-077125, Romania

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ABSTRACT

Radiological impact models are important tools that support nuclear safety. For tritium, a special radionuclide that readily enters the life cycle, the processes involved in its transport into the environment are complex and inadequately understood. For example, tritiated water (HTO) enters plants by leaf and root uptake and is converted to organically bound tritium (OBT) in exchangeable and non-exchangeable forms; however, the observed OBT/HTO ratios in crops exhibit large variability and contradict the current models for routine releases. Non-routine or spike releases of tritium further complicate the prediction of OBT formation. The experimental data for a short and intense atmospheric contamination of wheat are presented together with various models' predictions. The experimental data on wheat demonstrate that the OBT formation is a long process, it is dependent on receptor location and stack dynamics, there are differences between night and day releases, and the HTO dynamics in leaf and ear is a very important contributor to OBT formation.

1. Introduction

Tritium (^{3}H) is present in the environment as a result of both natural and anthropogenic sources. Large quantities of tritium are currently produced in heavy water reactors and fuel reprocessing plants, and it is anticipated that the development of fusion energy will increase environmental releases. Romania develops nuclear energy using Canadian heavy water reactors and two CANDU 6 units are in operation having large tritium loads. The past and present Romanian research studies cover environmental tritium monitoring (Paunescu et al., 2002), environmental tritium modelling (Barry et al., 1999), and it was pointed out that tritium has to be considered as a special radionuclide (Galeriu and Melintescu, 2010). Tritiated water (HTO) and tritiated gas (HT) are the major forms initially released in environment and the organic forms (tritiated methane or tritiated formaldehyde) have minor releases (Amano, 1995; Kakiuchi et al., 2002).

In the process of photosynthesis, plants produce organic matter using solar light as energy source, from carbon dioxide from the air, nutrients from soil, and water from soil or air. Because tritium is larger than hydrogen, the organic forms of tritium are produced less readily than the organic forms of hydrogen. The organic forms of tritium are generically called organically bound tritium (OBT). Following the definitions of Kim et al. (2013), there are three types of OBT: exchangeable OBT, non-exchangeable OBT, and a special form of buried tritium (*i.e.* tritium included in the hydration shell of biomolecules). How stable tritium is within such organic compounds depends on the nature of the bond between tritium and the organic molecule and on the organic molecule affinity with the different biological tissues (ASN, 2010). When tritium is bound to oxygen, sulphur or nitrogen, it can be easily exchanged with tritium in the HTO (or H_2O) and the exchangeable organically bound tritium (E-OBT) is formed. When tritium is covalently bound to carbon, only enzymatic reactions can destroy the bound and non-exchangeable OBT (NE-OBT) is formed. Buried tritium, which is inaccessible because of the physical structure of the organic molecule, quickly exchanges with hydrogen atoms in the body following digestions and, consequently, it increases the amount of tritium in the body water. The time when tritium remains incorporated therefore depends on the biomolecular turnover: fast, in the case of molecules involved in the energy cycle and slow, in the case of structuring molecules or macromolecules such as DNA or energy reserve molecule. Due to longer residence in the organism, NE-OBT is of first concern for health effects of a radiological dose.

* Corresponding author.
E-mail addresses: ancameli@ifin.nipne.ro, melianca@yahoo.com (A. Melintescu).

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The methods to measure the HTO in environmental samples are well established but the OBT measurements are expensive and difficult. OBT measurements in all food chain components (human and non-human) are not possible. The alternative is to use radiological/environmental impact assessment models (RIA/EIA). These models can be defined as research grade and decision making models. The latter are used in design, licensing, normal operation, accident prevention and management. The decision making models must meet the following requirements:

- Relatively simple;
- Transparent;
- Easy to program;
- Results should be conservative, yet reasonable;
- Deterministic calculations possible (worst case assessments);
- Probabilistic calculations possible (95% percentile as worst case).

Finally, when the models are applied in operational context, they must quickly provide results (*i.e.* have a short run time).

During the working group meeting dedicated to "Tritium Accidents" (WG 7) of Environmental Modelling for Radiation Safety (EMRAS II) programme coordinated by International Atomic Energy Agency (IAEA) (http://wwwns.iaea.org/projects/emras/emras2/working-groups/working-group-seven.asp?s=8), one of the questions was if it is possible to have such models for tritium, due to high complexity of processes involved in the environmental tritium dynamics. A robust model for accidental tritium releases must minimize the uncertainty that can arise from the model structure, model parameters, and those situations that need special attention (see Chapter 15 in IAEA, 2014a). For routine releases of heavy water reactors, a steady state model developed by the Canadian Standard Association (CSA, 2014a) is generally used. Recently, claims have been made that the CSA model is not conservative with regard to OBT concentration in food (Thompson et al., 2015). Romania uses both Canadian (CSA, 2014a) and European practice (https://ec.europa.eu/energy/en/topics/nuclear-energy/radiation-protection) in its radiation protection programmes and some differences between those practices were noted (Galeriu et al., 2009a). The anti-nuclear groups claimed that the current models for routine release largely under-estimate the public dose because it is based on the yearly average of the air concentration and ignores spikes in the releases (Fairlie, 2010). Indeed, this claim is justified because at the receptor, the air concentration fluctuates and the equilibrium conditions are not reached as CSA (2014a) considered. In the extreme situation of a short and intense emission of tritium, a detailed analysis of the uptake processes is still needed, in order to provide a robust prediction of OBT production in crops at harvest. An analysis of the present state of knowledge regarding OBT modelling in plants during normal operation of nuclear utilities, as well as spike releases of tritium emissions are provided in the present study in order to support the current efforts regarding the tritium model improvements (Melintescu et al., 2015; Galeriu and Melintescu, 2016).

2. Materials and methods

2.1. Uncertainty of OBT/HTO ratios for routine releases

Pressurized heavy water reactors (PHWR) were developed in Canada and have higher tritium emissions than other energetic reactors. For calculation of Derived Release Limits (DRL) for assessment of the public dose during normal operation of PHWRs, the standard guide used in Canada and Romania in the past ignored the OBT contribution to the food chain (CSA, 1987). Subsequently, the importance of OBT was pointed out in relation with fusion research (Murphy, 1993) and the consideration of OBT contribution to the food chain was proposed soon afterwards (Galeriu, 1994). The contribution of OBT was considered in the second revision of the Canadian guide (CSA, 2008). The last revision of the guide (CSA, 2014a,b) is based on the same specific activity (SA) approach as the previous guides, assuming full equilibrium in all environmental compartments (CSA, 1987, 2008). The SA approach is also used by IAEA in its coordinated research studies (IAEA, 2009, 2010) and details regarding the CSA (2014a) and IAEA (2009, 2010) approaches are given in Appendix A. Based on simplifying assumptions of SA approach, the ratio between OBT concentration (measured by water of combustion), C_{OBT} and HTO concentration in plant water (leaves), C_{TFWT} is constant. The difference between those two approaches is that CSA considers the total OBT in plant and the isotopic discrimination factor, ID_p has a recommended constant value of 0.8 (range of 0.64–1.3), while IAEA considers NE-OBT in plant and the partition factor for plants, R_p has a recommended constant value of 0.54 as a geometric mean (GM) and a geometric standard deviation (GSD) of 1.16. The data considered by IAEA (2009) were for barley, maize and alfalfa, grown in laboratory controlled experiments and had equilibrium values. The experimental values for the partition factor, R_p cover a range of 0.4–0.68. The partition factor, R_p includes both the isotopic discrimination factor (with an average value close to the ID_p given by CSA, 2014a) and the contribution of E-OBT to total OBT.

For public dose assessment during routine emissions and based on equilibrium assumption, both approaches consider the crop contamination at harvest and they ignore the losses due to storage and food processing. For the assessment of plant tissue free water tritium (TFWT), CSA (2014a) considers the air HTO concentration (with a reduction factor, RF_p due to the lower contribution of soil HTO), while IAEA (2009) separately considers the tritium transfer between air and plant (air pathway) and that between soil and plant (root pathway) (for details see Appendix A). The air HTO concentration is a yearly average or an average over the vegetation period. Disregarding the difference between total OBT and NE-OBT, the OBT/HTO ratio for equilibrium conditions is close to 0.7 with a range of 0.4–1.3.

Based on a detailed analysis of a large range of experimental values for OBT and HTO in agricultural products, it was observed that the ratio between OBT (water of combustion) and TFWT (HTO in leaf water) largely differs from the equilibrium value of 0.7 (CNSC, 2013; Korolevych et al., 2014; Thompson et al., 2015) with many values higher than 5 and few values higher than 10. Traditionally, this ratio is expressed as OBT/HTO, but in fact, OBT represents tritium concentration in water of combustion and HTO represents tritium concentration in plant/animal water, which is actually TFWT. Theoretically, that ratio should be expressed as OBT/TFWT, but for the sake of simplicity, the present study keeps the traditional expression of OBT/HTO ratio.

The uncertainty and under estimates of OBT concentrations come from the equilibrium assumptions based on a long term average of HTO concentration in air. In real field conditions, there is no equilibrium as it was previously pointed out (IAEA, 2008a). In fact, at the receptor where the measurements were carried out, there is a fluctuating HTO concentration in air, the HTO concentration in leaf has a diurnal variability and the OBT accumulation in different plant parts is a slow process. The sampling is usually carried out during the working hours and the HTO concentration in leaf can be low after the plume passage and high during the plume passage.

The HTO concentrations in plants reflect conditions in the few hours before sampling, while monitoring results are yearly or monthly. On the other hand, residence times for OBT in plants are