Scientific report – extended summary

July – December 2017

Open problems in radiological risk assessment of tritium emissions including climate changes PN-III-P4-ID-PCE-2016-0218 (OPTRAS)

Tritium (^3H) 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.

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).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. When tritium is bound to oxygen, sulphur or nitrogen, it can be easily exchanged with tritium in the tritiated water (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.

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).

Tritium dynamics in crops as both HTO and OBT follows the water cycle and dry matter dynamics, respectively. The plant characteristic parameters in CROPTRIT model are taken from crop growth models including water. The crop growth models must be robust in relation to crops, soil and weather for the area of interest. The area of interest in CROPTRIT is Cernavoda NPP (Romania) operating two CANDU 600 reactors. The crops of interest are those cultivated in Cernavoda area such as: winter wheat, rye, corn, sunflower, grapes, pasture and vegetables, but the model is restricted to a single crop, winter wheat. The soils are mostly sand and silty with a predominant loamy texture. The climate in Cernavoda area is quasi-dry with an average precipitation of about 400 mL y^{-1} and with an average evapotranspiration of about 700 mm y^{-1}. The growth sub-model in CROPTRIT is an adaptation of WOFOST and ORYZA models to Romanian conditions.

The HTO vapour transport from air to soil depends on the concentration gradient between air and soil surface and on the exchange velocity between air and soil.

The concentration of HTO in the plant water depends on the concentration gradient between air and the stomata cavities and on the exchange velocity between air and plant canopy. Depending on the HTO flux direction, deposition (HTO flux from air to plant) or reemission (HTO flux from plant to air) takes place. CROPTRIT model considers only tritium deposition on plants and soil.

In many studies regarding the water (H_2O) transfer in plants, the so called “big leaf” approach was abandoned for the dual source model, which makes a better distinction between the plant canopy and soil, as well as between the leaves transpiration and soil evaporation. The dual source model (Shuttleworth and Wallace, 1989) was adapted for tritium (Melintescu and Galeriu, 2005) and is used in CROPTRT model for calculating the evapotranspiration. The dual source model needs to know the net radiation and that absorbed by soil and consequently, the equations for various conductances and fluxes are more complicated.

For the atmospheric pathway, the modelling of exchange velocity together with canopy conductance and leaves temperature (coming from the energy balance) are needed. A special attention is given to the stomatal conductance.

For root pathway, the HTO concentration in transpiration water is needed, and consequently, the evolution of HTO concentration in soil profile and rate of water extraction by root are needed.

Once known the HTO concentration in leaf, the rate of OBT production can be assessed.

CROPTRIT model needs the assessment of the hourly plant evapotranspiration. The classical approach (FAO, AQUACROP) for the assessment of evapotranspiration considers a reference crop, a crop factor (specific to each crop and depending on plant development stage) and a constant day / night stomatal resistance and is not considered in CROPTRIT model, because realistic day and mostly night canopy conductance are needed. Consequently, CROPTRIT model considers the dual source approach (complex approach) for the assessment of evapotranspiration (Shuttleworth and Wallace, 1989). The dual source approach assessing the net solar energy and that absorbed by soil needs the solving equations for energy and water mass conservation at soil – vegetation interface together with solving the dynamic equations for water-vapours-temperature. A coherent approach of the dynamic transfer of tritium in air-plants-soil continuum needs the concomitant solving of all equations and boundary conditions. The constant temperature in soil together with HTO vapours transfer in soil are considered. At the beginning of each hourly interval, the sub-model for soil transpiration based on fast numerical solution (Ross, 2003) is run. The resulting water stress, as well as real transpiration is introduced in the sub-model developed for the assessment of HTO dynamics in plants and that for OBT production. At the end of each day, the sub-model for assessment of dry matter and OBT production and its partition to various plant parts is run. COPTRIT model is run with hourly meteorological data starting with plant emergence until plant maturity and harvest.

Plants regulate their stomatal conductance in order to optimize the balance between carbon uptake and water loss. A major limitation of this process is the rate at which stomata open at light or close at dark or as a consequence of water deficit in soil. In favourable conditions of low evaporation and intense light, the high limitation of CO_2 assimilation rate is determined by the maximum value at which the stomatal conductance, g_{op} operates. In case of a severe water deficit coming from high evaporation necessities and/or dry soil, plants survive by complete close of their stomata and by leaf cuticles impermeable to water, in order to minimise their water loss. Stomata close at dark and stay closed for almost all night long, but often, the stomata closure is not complete. In fact, the nocturnal conductance is enough to allow a significant transpiration rate. Transpiration rate at night represents 3 – 5 % from that at day, but in arid and semi-arid regions it can be higher than 30 %.

At the canopy level, stomatal conductance is integrated on leaf area index (LAI), considering the simplifying assumption that the CO_2 concentration at leaf surface, C_s and water vapour pressure deficit at plant level, D_s are constant.

In order to underpin the hypotheses of CROPTRIT model, a literature study was carried out to emphasise the role of mesophyll conductance, the minimum stomatal conductance, as well their dependence on plant type. Generally, D_* significantly depends on temperature, minimum stomatal conductance and mesophyll conductance (Fig. 1).

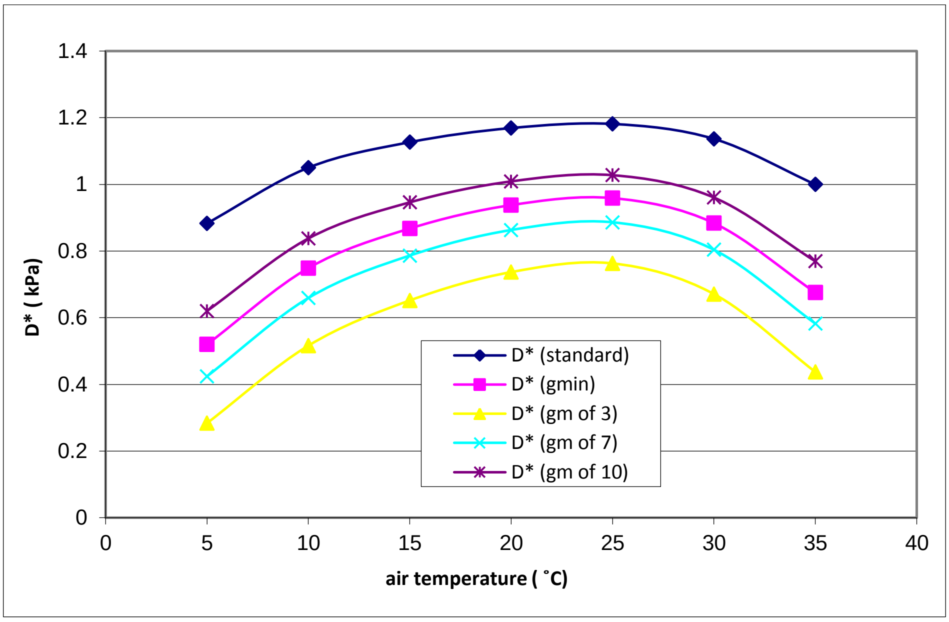

Fig. 1. Dependence of vapour pressure deficit for closed stomata on air temperature, minimal conductance and mesophyll conductance

The soil sub-model in CROPTRIT was tested for a number of soil layers needed for an acceptable precision, for the soil type and its properties and for the influence of root characteristics. In case of a root depth of 1 m and a soil depth of 2 m, the model performances are very good. The conservation of water mass and tritium activity can be obtained for 14 soil layers and the calculation time is less than a second for o sequence of 70 days.

The soil sub-model was tested for HTO dynamics in soil and leaves for a dry and a wet contamination coming from an intense rain. An extreme tritiated event was assumed when the air contamination with HTO sharply varies from background value of 100 Bq L^{-1} as in field conditions in Romania and Canada up to values of 5.9 x 10^6 Bq L^{-1} (about 6.5 x 10^4 Bq m^{-3}) (low effective plume height), for 6 hours, in the morning, with clear sky. That represents a dry deposition on soil and plants. For wet deposition, it was assumed that for the whole interval, a precipitation of 25 mm h^{-1} occurred, with a total of 150 mm h^{-1} (an extreme case), at 1 km away from the emission source, the effective release height was 30 – 60 m from soil, atmospheric stability class D (neutral) and wind velocity of 4 m s^{-1}, with wind blowing in the same direction. That is considered the worst case scenario. The tritiated event occurred one month before anthesis. Crop considered is irrigated wheat with clean water of 24 mm d^{-1} at days 15, 30, 45, and 60, respectively. The event was simulated between harvest and 45 days after anthesis. The reference PET was 5.5 mm d^{-1}, the actual PET was corrected for crop coefficient and the potential transpiration was 0.75 of PET. All the days have the same diurnal pattern for PET (clear sky, excepting the rain event). LAI was 5.5 from day 0 up to day 50 and decreases at 2 until harvest. Realistic root depth of 1 m and root profile were considered, together with a soil column of 2 m, compensating the root uptake. The HTO concentration in rain water is assessed at 5.8 x 10^{5} Bq L^{-1}. During rain event, the sky is clouded, the air temperature decreases and consequently, the plant canopy conductance decreases. The plant considered in CROPTRIT is wheat, with hydrophobic leaves and for an intense rain, only the inferior layers of leaves can accumulate a water layer. Consequently, the canopy conductance in case of wet deposition is reduced to a half of that for dry deposition. For dry deposition, the soil is contaminated in a superficial layer and the HTO dynamics at deeper soil layers is slow. For the first 350 hours, the HTO dynamics in 10 soil layers with thickness of 1.5 cm is given in Fig. 2.

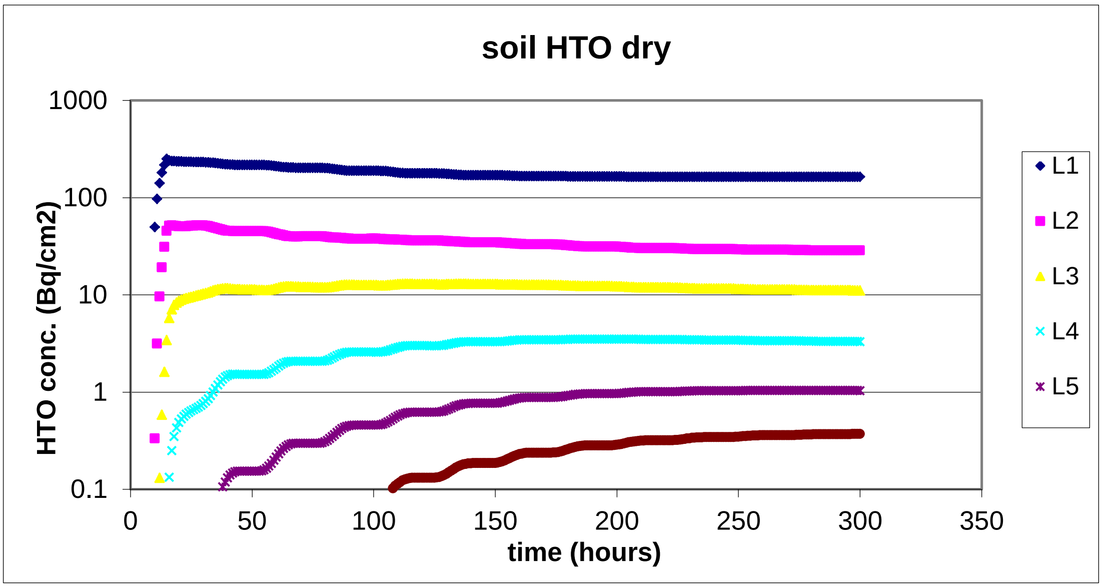

Fig. 2. HTO concentration in different soil layers for dry deposition

For a precipitation event, a large quantity of HTO is deposited on the soil together with a large quantity of water. The HTO dynamics is very fast and is given in Fig. 3. The sixth soil layer (L6 in Fig. 3) is contaminated in the first 10 hours contrary to the case of dry deposition (L6 in Fig. 2) where contamination starts at hour 106 (day 4). The HTO dynamics in the first two layers (L1 and L2 in Fig. 3) is not satisfactory modelled because the temperature and water vapours effects are not considered, but their contribution to transpiration flux is reduced.

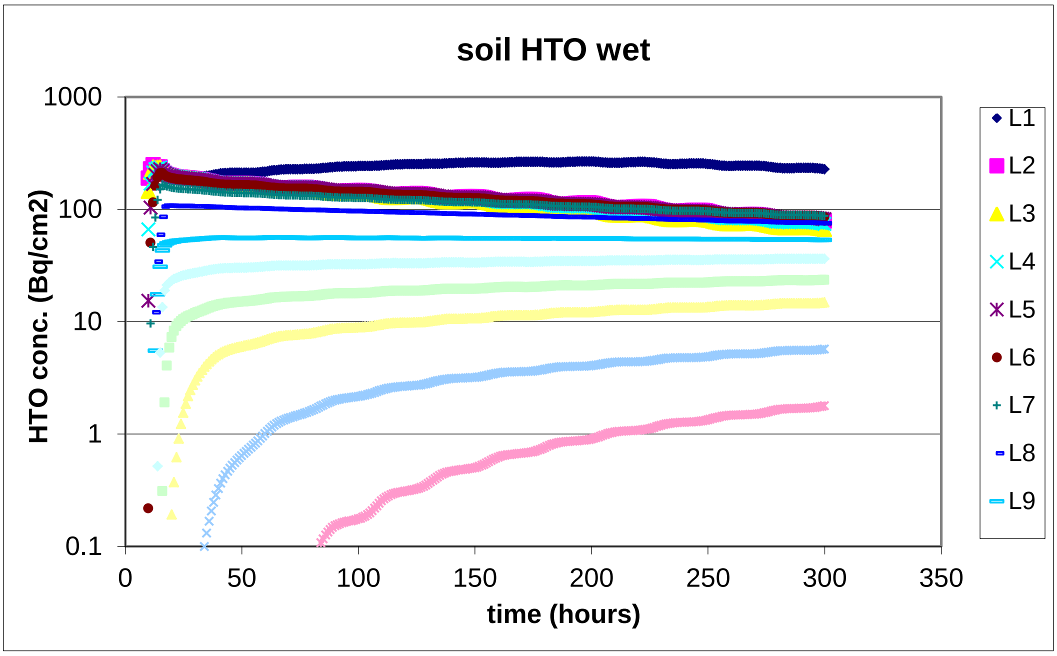

Fig. 3. HTO concentration in different soil layers for wet deposition

For OBT production at night, first model tested with experimental data was recently published (Galeriu and Melintescu, 2017).

The model was first applied for wheat, where many experimental data are available for controlled conditions. The best information on respiration and cultivar characteristics were considered but protein turnover was ignored. The photosynthesis model was tested to be appropriate with the experimental biomass dynamics and the short term variability of meteorological data. The night OBT production depends on the starch degradation rate and leaf HTO concentration. The numerical experiments started with the wheat starch degradation rate and involved the variation of leaf HTO concentration during the night time. A first case uses the default sub-model for leaf HTO with a background of 100 Bq L^{-1}, as in field conditions in Romania and Canada. The dynamics of leaf and grain OBT (combustion water) was normalized by the leaf HTO concentration at the end of exposure (TLI). Comparing the model results with the experimental data in controlled conditions, an over-prediction with a factor of 4 for leaf OBT (few hours after the end of exposure) and a factor of 5 for grain at harvest respectively, was observed. Then, a low background for leaf HTO (as in the controlled experiment) was considered and an under-prediction with a factor of 3 was observed. The experiment was carried out in conditions of low wind speed, but the wind was not measured. The experimental data for leaf HTO concentration were used and the model results for grain were very close with the experimental data in the first day (Fig. 4) with an error of 30 % at harvest.

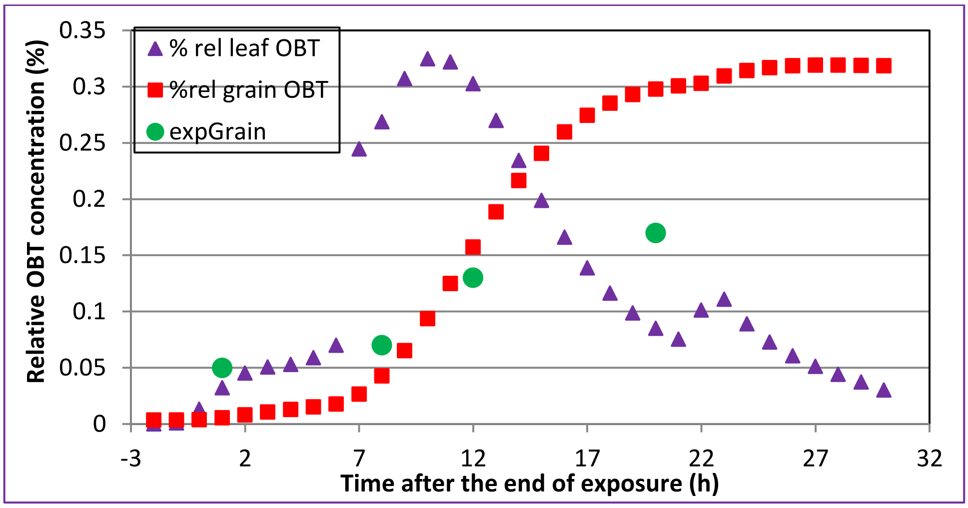

Fig. 4. Relative OBT concentration in leaf and grain of wheat in the first 28 hours after the night exposure of one hour starting at 23:00

In the same conditions, the starch degradation was varied from the wheat case to linear case or using the data for maize. Miss-predictions were at a factor of 2 to 3. Similar results were obtained for rice. It can be noted that the model uncertainty due to leaf HTO is larger than that for starch degradation. Recent advances in crop eco-physiology must be used, because they show a correlation between night respiration and night transpiration (crop adaptation for saving both carbon and water use). In the case of Romanian CANDU reactors, preliminary results show no excessive contamination in the night for wheat, barley, and maize compared with day conditions.

For leaf, the protein turnover is important and uses about 20 % from the sucrose compartment after anthesis, decreasing to maturity. Based on little experimental information, the dynamic of labile protein and carbohydrates can be reproduced for wheat, but for tritiated compounds further research is needed to assess the water addition coefficient. This is important mostly for hay and leafy vegetables containing 10-20 % dry matter.