Scientific report – extended summary

January – December 2019

Open problems in radiological risk assessment of tritium emissions including climate changes PN-III-P4-ID-PCE-2016-0218 – IDEI 191/2017 (OPTRAS)

The upgrades of a dynamic operational model for tritium transfer (as HTO and OBT) in crops (CROPTRIT) together with model tests and validation with the available experimental data for winter wheat were presented in the Reports for 2017 and 2018. The selected crops are specific to Cernavoda NPP site (Romania) operating two CANDU 6 reactors where winter wheat is of first concern, then pasture, maize, and wine grapes (order of planting). The full data base of Romanian agricultural research was accessed. Night processes involved in OBT production are considered in the upgraded CROPTRIT model. In the present report, CROPTRIT was tested for its uncertainty and sensitivity analysis, the model was adapted for Cernavoda NPP as an operational tool (parsimonious modelling) and the model for dynamic transfer of tritium and radiocarbon to biota was upgraded. The model application to radiocesium was also carried out.

The term of uncertainty refers to the lack of knowledge or information regarding the modelled processes, parameters (which vary) or model constants, input data and concepts on which the model is based.

Quality assurance of a model could be defined as a complex of protocols and guidance for correct application of models by using the optimal methods (best practice), assuring the consensus between participants (including the users) and certifying that model performance is according to project goal.

Uncertainty affecting the quality of the model has certain dominant components:

- Conceptual uncertainty (epistemological) due to incomplete knowledge of some processes, to unjustified simplifications or some limitations regarding spatial or temporal resolution;
- Technical uncertainty of some model quantities, such as input data or model parameters, measurement errors or parameters aggregation;
- Niche uncertainties coming from model application outside the range for which it was developed or by combination of many models with different spatial and temporal scales.

For conceptual uncertainty, there is a list of processes not well understood such as:

- Interception of tritium wet deposition by plants and its uptake:

- Tritium interception by bare and vegetated soil;
- HTO transfer and its dynamics in soil-vegetation-atmosphere continuum;
- HTO reemission from soil as a secondary tritium source;
- OBT formation during the night time;
- OBT oxidation during food processing and storage;
- OBT cycling in dead vegetation deposited on soil and OBT cycling in soil;

Another important issue is models inter comparison and their tests with experimental data for:

- Dry and wet tritium deposition after dry and wet seasons:

- Dry and wet deposition of tritium at day and night;
- Dry and wet deposition of tritium during various seasons.

The major uncertainty of any tritium model, as well as CROPTRIT, is represented by the current knowledge of OBT formation in plants in case of normal and accidental release of HTO from nuclear facilities. In the following, a brief presentation of the uncertainty sources coming from OBT formation is carried out.

For routine emissions at CANDU reactors (Canada, Romania) the dose assessment to population is based on the Canadian Standard, where the OBT concentration in agricultural products (combustion water) is in a fixed ratio with HTO concentration in leaves. OBT/HTO ratio is based on laboratory experiments, where all compartments (air, water, soil, plant) are in equilibrium. Analysing a large experimental data base, including monitoring results, it is found that this ratio has a large range, between 0.1 and more than 10, too large comparing to the equilibrium value of 0.7. Recently, the Canadian Standard was criticized and a clarification is needed. In field conditions, the HTO concentration has a large variability, depending on meteorological conditions during emission. Each plant type has its own growth dynamics and consequently, OBT and HTO concentration at harvest depends on plant type, receptor location, meteorological conditions and emission dynamics at stack. In practice, the equilibrium conditions are never reached. Based on experimental observations from Wolsong NPP (CANDU reactors in South Korea), it is observed that OBT/HTO ratio is specific to the location, year and plant (Figure 1) and varies depending on year and sampling time.

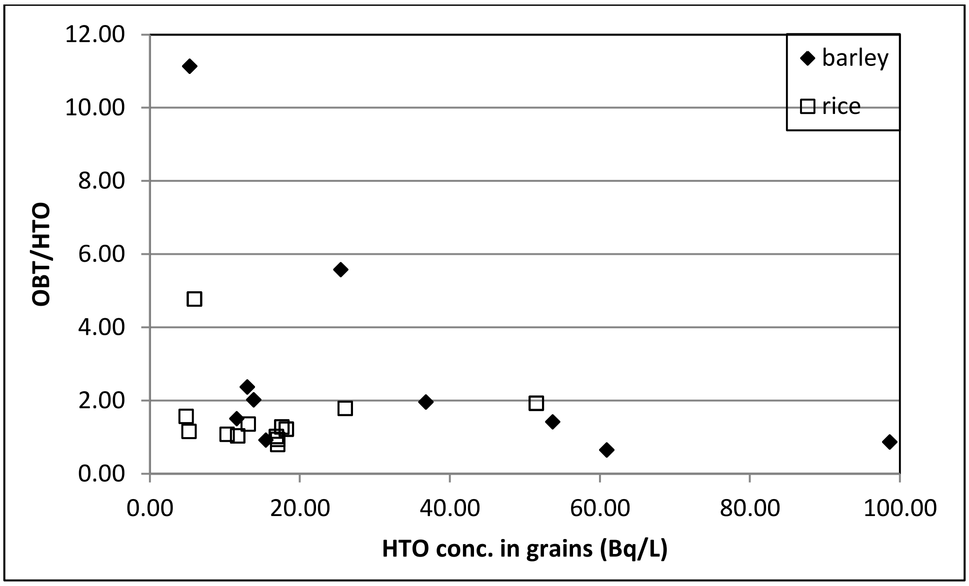

Figure 1. OBT/HTO ratio in barley and rice

For cereals (barley and rice), the average experimental ratio is about 2, for cabbage is 0.45, for persimmon is 0.58. For cereals, the equilibrium value is exceeded. For milk, the Canadian Standard gives a value of 0.25-0.4 for the OBT/HTO ratio, but the experimental data have an average of 1.2. The Canadian Standard is based on the following relationship:

$$C_{OBT} = DW_p * ID_p * WE_p * C_{TFWT}$$

where: RF_p is the reduction factor used because tritium concentration in soil is much smaller than that in air (unit less); DW_p is the dry matter (dm) fraction of plant (kg dry weight (dw) plant kg^{-1} fresh weight (fw) plant); ID_p is the isotopic discrimination factor due to plant physiology (unit less); WE_p is the water equivalent of organic matter (at combustion in OBT measurements); H_a is the absolute atmospheric humidity (L m^{-3}); C_{air} is the HTO concentration in air (Bq m^{-3}); C_{am} is the HTO concentration in air moisture (Bq m^{-3}); C_{TFWT} is the HTO concentration in the leaf free water (Bq L^{-1})

The recommended value for ID_p by the Canadian Standard corresponds to the equilibrium value, but we propose a probabilistic assessment and recommend a range of values (with a distribution probability) for all Canadian model parameters.

The proposed probabilistic assessment is done for Cernavoda NPP, considering the parameters variability and the necessity of local values (or probabilistic values) and not the generic ones (Tables 1 – 3).

Table 1. Public dose (Sv $year^{-1}$) coming from different tritium contamination pathways for children of 1 year and 10 years old and for an adult when OBT/HTO ratio is 0.7 and tritium concentration in animal drinking water is 4.5 Bq L^{-1}

Contamination pathway	*1 y*	*10 y*	*Adult*
Inhalation	1.97×10^{-7}	2.71×10^{-7}	2.27×10^{-7}
Drinking water	8.67×10^{-8}	5.84×10^{-8}	7.68×10^{-8}
Food HTO	6.17×10^{-7}	4.29×10^{-7}	3.32×10^{-7}
Food OBT	9.08×10^{-8}	9.79×10^{-8}	8.71×10^{-8}
Total	9.92×10^{-7}	8.57×10^{-7}	7.23×10^{-7}

Table 2. Public dose (Sv $year^{-1}$) coming from different tritium contamination pathways for children of 1 year and 10 years old and for an adult when OBT/HTO ratio is 0.7 and tritium concentration in animal drinking water is 40 Bq L^{-1}

Contamination pathway	*1 y*	*10 y*	*Adult*
Inhalation	1.97×10^{-7}	2.71×10^{-7}	2.27×10^{-7}
Drinking water	8.67×10^{-8}	5.84×10^{-8}	7.68×10^{-8}
Food HTO	1.08×10^{-6}	5.76×10^{-7}	4.48×10^{-7}
Food OBT	1.39×10^{-7}	1.15×10^{-7}	9.87×10^{-8}
Total	1.50×10^{-6}	1.02×10^{-6}	8.50×10^{-7}

Table 3. Public dose (Sv $year^{-1}$) coming from different tritium contamination pathways for children of 1 year and 10 years old and for an adult when OBT/HTO ratio is 10 and tritium concentration in animal drinking water is 4.5 Bq L^{-1}

Contamination pathway	*1 y*	*10 y*	*Adult*
Inhalation	1.97×10^{-7}	2.71×10^{-7}	2.27×10^{-7}
Drinking water	8.67×10^{-8}	5.84×10^{-8}	7.68×10^{-8}
Food HTO	6.17×10^{-7}	4.29×10^{-7}	3.32×10^{-7}
Food OBT	1.05×10^{-6}	1.19×10^{-6}	1.07×10^{-6}
Total	1.95×10^{-6}	1.95×10^{-6}	1.70×10^{-6}

It can be observed that the uncertainty introduced by OBT/HTO ratio is comparable with that introduced by tritium concentration in animal drinking water and the conservative dose for the public is only μSv y^{-1} for an air HTO yearly average concentration of 1 Bq m^{-3}.

The anti-nuclear groups claimed that the public dose is largely under estimated because it is based on the yearly average of the releases, disregarding the short time higher releases. The National Dose Assessment Working Group (NDAWG) (UK) considered in detail the case of a short-term and intense emission comparable with the same total annual emission. It was considered the same total emission distributed on a short period (a day or less) or distributed during a year. For spike release, three cases were analysed: a) realistic - release during a normal day over 12 hours in neutral meteorological conditions (it can include rain and moderate wind direction changes, in conformity to local multiyear data); b) cautious - release of 30 minutes in neutral meteorological condition (class D), wind direction through production area, including rain; c) cautious - 30 minutes release in stable meteorological condition (class F), wind over production area, no rain. For continuous release over a year, the average UK weather was considered. The considered tritium release is 1 TBq for the whole year (continuous) or for the spike release. We used UFOTRI and CROPTRIT models for this case (Table 4).

Table 4. The ingestion doses (μSv $year^{-1}$) for infant, child and adult for an atmospheric tritium emission of 1 TBq for various atmospheric stability conditions and durations

Case	*Normal day*	*Category D*	*Category F*	*Continuous release, 1 year*	*Category F/continuous release ratio*
Infant	1.33	2.22	8.24	0.23	35.21
Child	1.68	2.77	10.20	0.24	41.80
Adult	1.80	3.96	14.30	0.26	55.21

In Table 4, it is observed that the spike releases gives higher public doses, but those value could not be definitely quantitatively accepted, because all the models do not have yet the predictive power for robust assessment.

The controlled experiments of wheat contamination with air HTO were analyzed and compared with model results in an inter comparison exercise coordinated by IAEA. We reanalyzed the results and their interpretation based on two facts:

1) The empirical relationship between OBT concentration in grains at harvest and HTO integral concentration in leaf and ear:

$$C_{OBT-grain} = 0.48 * [IC_{leaf-day} + 0.2 * IC_{leaf-night} + 0.5 * \left(IC_{ear-day} + 0.2 * IC_{ear-night}\right)]$$

The empirical relationship, checked by us, pointed out the difference between OBT production between day and night (a factor of 0.2 for night), but the ear contribution too, to OBT production (photosynthesis in green ear).

In our model, the ear contribution to photosynthesis is introduced by the increasing of leaf area index.

2) The dynamics of OBT/HTO ratio after the end of exposure of an hour and for the experiments at various hours of the day (Figure 2)

In Figure 2, it is observed that OBT/HTO ratio has low values at the end of each exposure and gradually increased up to 10 after 24 hours, demonstrating that the OBT formation is a complex and long process and the OBT/HTO ratio strongly depends on the sampling time after exposure.

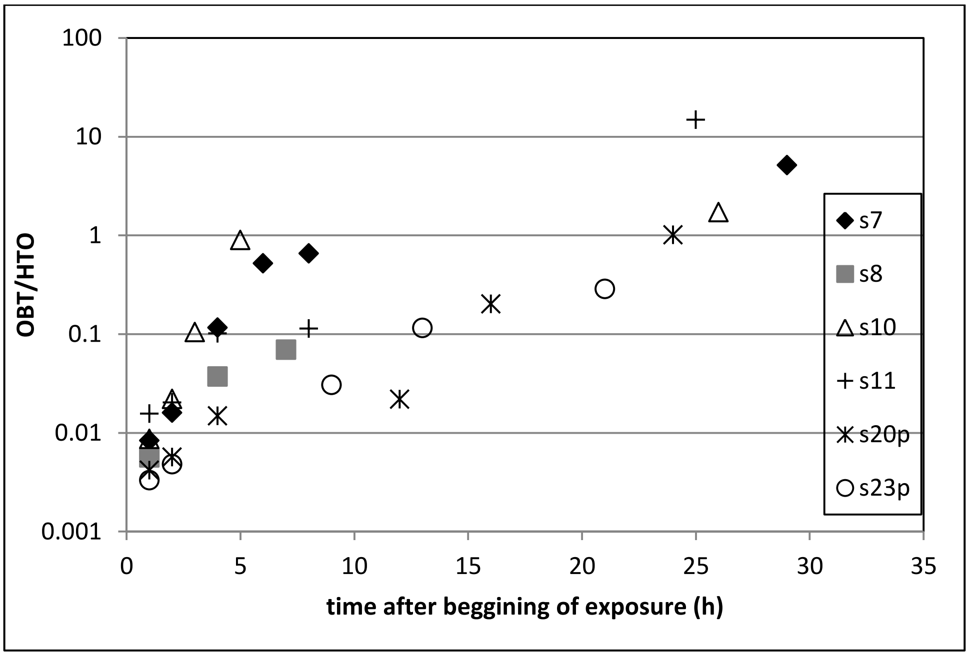

Figure 2. Experimental OBT/HTO ratio for wheat leaves at various exposures carried out at different hours of the day and night (s7, s8, s10, s11, s20p, s23p)

The increases of public dose due to routine emissions cannot be ignored and they are due to non-equilibrium processes in field conditions. If the increases in routine emissions coincide with the period when most of the agricultural plants around the NPP are one month before harvest, the food chain contamination may be higher than for uniform emissions (the same emission as for spike releases) during a year. For continental areas, there is no plant growth during winter time. The uncertainty of the current dynamical models does not allow a quantitative analysis.

Due to the complexity of processes involved in OBT formation and HTO dynamics, the dedicated studies are still necessary. The efforts must be oriented to a better understanding of balances between root and atmospheric pathway on HTO dynamics subjected to various environmental conditions and to the potential role of OBT formation during the night time.

In case of CROPTRIT application to the operational case of Cernavoda NPP, we run the model for a situation when 30 g of HTO (11 PBq) are emitted in an hour, from a height of 50 m, stability class F, and wind speed of 1 m/s. That is not a maximal case, but an example. The total integrated concentration (TIC) at 1 km away from NPP is $0.7x10^{11}$ Bq s/m^3, 4-5 times lower than the recommended one. For this case, the public dose at 1 km away is 30 mSv and at 2.5 km away is 8 mSv (Figure 3).

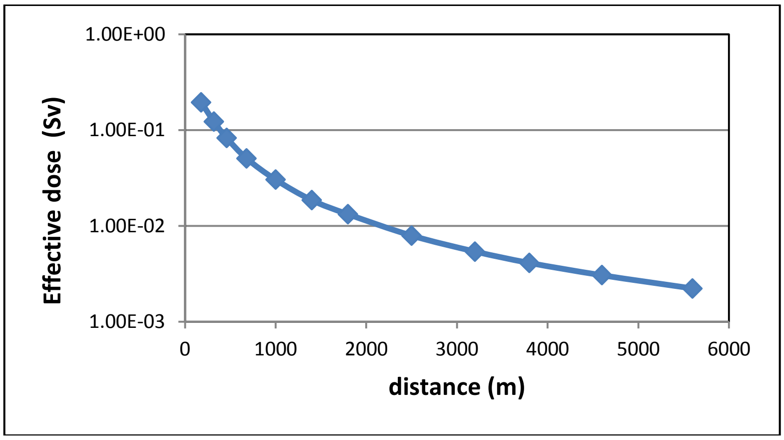

Figure 3. Dose dependence of distance for the most exposed person

HTO and OBT dynamics in the first 70 hours, acute phase, at 2.5 km away from the emission source is given in Figs. 4 and 5.

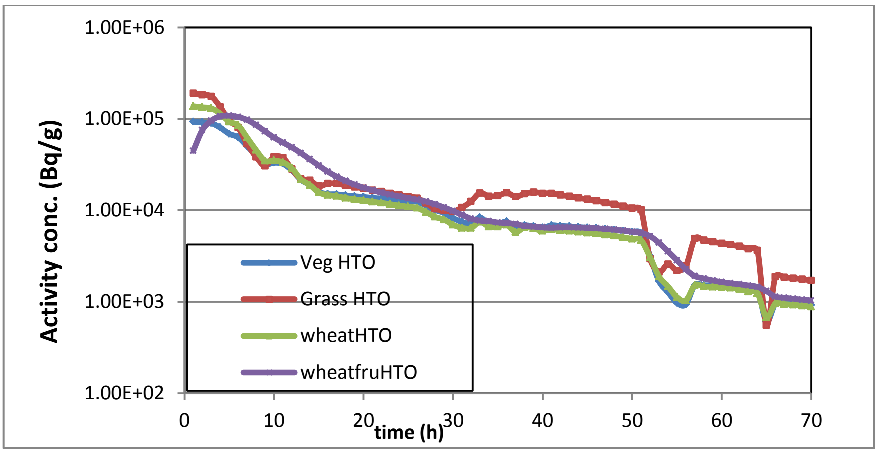

Fig. 4. HTO concentration in vegetation leaves, grass and wheat and in wheat grains

There is an abrupt decrease of HTO concentration after the plume passages, with values of a hundred times lower. The OBT concentration increases drastically in the first 10 hours, it decreases a bit the next day, and then remains constant until the hour 70 (Fig. 5).

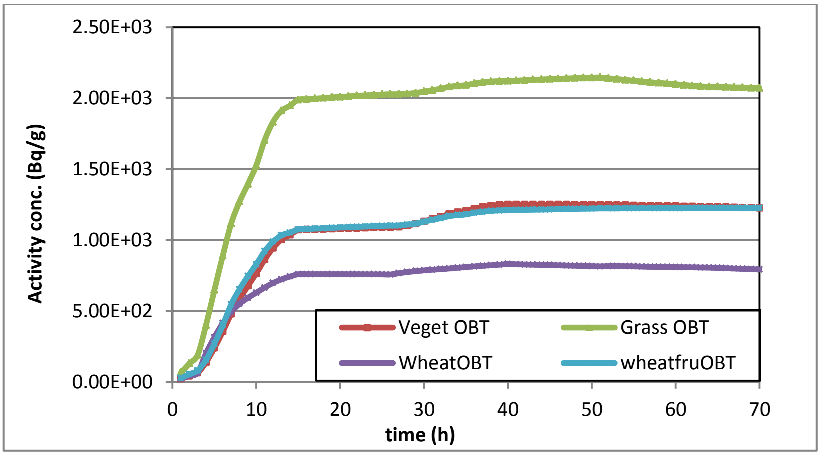

Fig. 5. OBT concentration in vegetation leaves, grass and wheat and in wheat grains

OBT dynamics in the first year at 2.5 km away from the emission source is given in Fig. 6. After the first days, the dynamics of OBT concentration is different in leaves comparing to other edible parts of the plant.

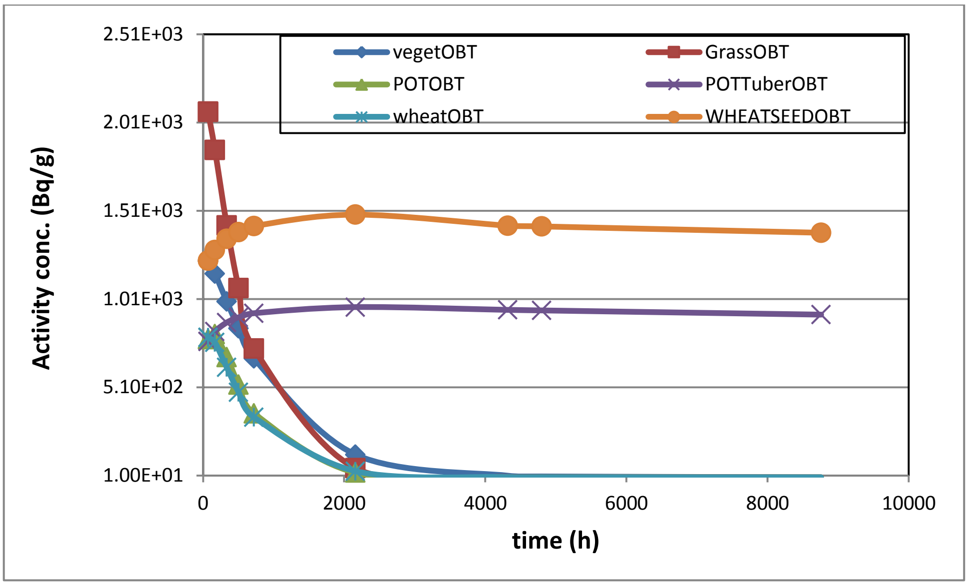

Fig. 6. Dynamics of OBT concentration in plants in the first year after the accident at 2.5 km away from the emission source

In leaves (vegetables, grass, wheat or potatoes), the OBT concentration significantly decreases in the first week and slowly thereafter (but constantly stays higher than the HTO concentration). The leaves are the primary source of OBT production, from where the OBT partition to other plant organs starts. Consequently, the OBT concentration in potato tubers and wheat grains increases in the first week ant stays almost constant until the harvest.

For night emissions, many exercises were carried out and we chose the maximal case, emission class F, at 20 m height, wind speed lower than 1 m/s. The maximum dose at 1 km away from the emission source is 46 mSv/year and 2.5 km away is 12.4 mSv/year, significantly higher than during the rain time (Table 5). The ingestion prevails with more than 77% and TIC at 1 km away is $1.3x10^{12}$.

Table 5. Dose variation with distance, percentage of dose coming from inhalation and ingestion and air integrated concentration (TIC) (night emission)

DISTANCE (m)	IH (%)	IG (%)	IHR (%)	EDE (Sv)	TIC (Bq s/m^3)
180	20.32	79.6	0.08	2.09E-01	6.2E+12
320	19.9	79.82	0.28	1.53E-01	4.46E+12
460	19.65	79.96	0.39	1.14E-01	3.27E+12
680	19.58	80.02	0.41	7.51E-02	2.16E+12
1000	19.37	80.15	0.47	4.59E-02	1.3E+12
1400	18.99	80.41	0.6	2.88E-02	8.02E+11
1800	18.72	80.57	0.71	1.99E-02	5.46E+11
2500	22.25	77.07	0.69	1.24E-02	4.05E+11
3200	23.22	75.99	0.79	8.12E-03	2.76E+11
3800	24.52	75.13	0.35	8.15E-03	2.93E+11

4600	24.55	75.09	0.35	4.09E-03	1.47E+11
5600	24.96	74.84	0.2	4.30E-03	1.57E+11

Based on our calculations, the potatoes and milk consumption is forbidden. The agricultural consumption on a surface of 50 km^2 exceeds $20x10^6$ kg and is compromised. There is a seasonal dependence of dose after an accident. For the daytime emission, CROPTRIT model includes all the agricultural products around Cernavoda NPP and gives a maximum of the dose during the autumn time (Fig. 7).

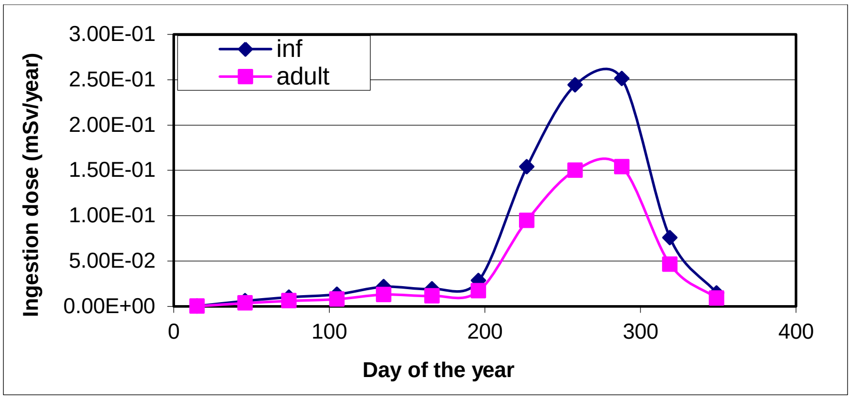

Fig. 7. Seasonal dependence of ingestion dose for a tritium accident

Summarizing, a tritium atmospheric accidental emission of 30 g (HTO) gives a significant radiological impact. In case of a favorable emission during the day time, clear sky, the public dose is 3-6 mSv/year and foodstuff contamination higher exceeds 10000Bq/kg. Countermeasures for food chain consumption are imposed for a long period of time (up to a year). For night time or rain emission, the consequences increase with an order of magnitude.

Any major nuclear facility must ensure the conservation of biodiversity regarding radiation protection of biota. A special concern is for tritium (^{3}H) and radiocarbon (^{14}C) transfer in wild mammals, birds and reptiles. Hydrogen and carbon are the main components of biological tissues and enter the life cycle. The present study improves the scientific bases of our previous model, analyses the uncertainty of input parameters and tests the model for a larger range of mammals and birds. The biological and metabolic half-times for organically bound tritium (OBT) and ^{14}C are linked with energy metabolism and recent results are revised in relation with metabolic scaling. A large data base regarding basal metabolic rate (BMR), field metabolic rate (FMR), and organ mass is used for input information of the present model, which considers brain as a separate compartment. Metabolic energy partition in organs of active animal is defined and the factors affecting the metabolic rate are analysed. Body and ambient temperature, diet and habitat, and phylogeny are important factors considered in animal adaptation to environment. The available experimental data for carbon turnover rates in animals are analysed and it is observed that the experimental conditions are not appropriate for wild animals. The link between $^{13,14}C$ and $^{134,137}Cs$ turnover rate is analysed and the present metabolic approach is successfully tested for mammals and reptiles. Considering animal adaptation and the large data base for $^{134,137}Cs$, the radiological impact of accidental releases of ^{3}H and ^{14}C on biota can be pursued in the future research.

The present study is based on a data base for wild mammals and birds derived from Navarrete et al. (2011) and Daan et al. (1990) and uses input information on basal metabolic rate (BMR) (*i.e.* minimal rate of energy expenditure by animals at rest), field metabolic rate (FMR), and organ mass for model application to a large range of animals all of which are relatively readily available in the literature. The present study uses energy metabolism and metabolic scaling knowledge to improve our earlier model (Galeriu et al., 2009; Melintescu and Galeriu, 2010).

A few simplifications considered in the present study must be pointed out:

- Basal metabolism is subjected to hormonal control and a representative value for a healthy adult of a species is based on measurements of many individuals of that species which is difficult to achieve in all situations;
- Mass of animals considered does not correspond with the mass of animal for which BMR was measured and the use of scaling relationships for mass may induce errors;
- Muscle mass was not measured in the experimental data base (Navarette et al., 2011) used in the present study and consequently, the muscle mass was assessed based on an extensive literature search which induces additional errors;
- The data base used to derive the allometric relationships for organs SpMR contains laboratory rat, humans and farm animals and do not contain wild animals and animals with mass lower than 200 g.

Based on those simplifications, the best BMR is reproduced with less than 30% in the normalised residuals. The cases with larger uncertainty (bolded in Table A1 of Appendix A) are discussed in the following. *Glaucomys volans* is a nocturnal species of flying squirrel which adapted its muscles and basal metabolism to its lifestyle. Its average resting metabolic rate (RMR) is higher during the winter time than during the summer time and its muscle mass is not well known. *Neomys anomalus*, *Neomys fodiens* and *Sorex araneus* are shrews with extremely high metabolic rates and food requirements and spend almost their entire life foraging. They cannot sleep or be inactive more than few hours per day and some of them enter torpor overnight. Consequently, it is difficult to achieve the condition of basal (standard) metabolic rate and their habit affects the organs SpMR in resting metabolism. They are all small mammals with mass lower than 25 g. The ratio between body surface and body volume of those small mammals is high and the heat loss from the body (at about 37.5 °C) to environment is high. Their lowest critical temperature is 25 °C and most of the time they are subjected to the cold stress, which involves a heat production from BAT and/or muscle shivering (Riek and Geiser, 2013). The SpMR of muscle can be higher than of other mammals with the same body mass. The data base used in the present study (Navarrete et al., 2011) do not provide information regarding the organs SpMR for small mammals and information provided for mouse (Martin and Fuhrman, 1955) is old and not yet confirmed. The specific information for small mammals is highly required and the present study tries to provide some details. Another source of uncertainty is the muscle mass. If the variability of the muscle mass between the measured and allometricaly calculated values is 20%, the overall effect on the variability of the calculated BMR is 8%.

The adaptation to ecological and climatic niche can modify the organ mass and organ SpMR. The muscle SpMR (basal metabolism) is important within the present model and improved data are needed.

Based on previous model (Melintescu and Galeriu, 2010) and present model, the maximum value of the viscera energy partition fraction we have assumed is too high and too little energy remains for muscle, which has a high contribution to the metabolism of birds. Flight requires a large amount of energy and muscle, lung and heart metabolism of birds is higher than that of mammals.

The organs SpMR of mammals are not appropriate for birds and in absence of any relevant information the uncertainty in energy partition to model compartments is high. In any potential application of the model for birds, the same average values as for mammals must be currently used, but with a higher uncertainty. The results for brain provided in the present study suggest its significant contribution to BMR of birds with body mass less than 150 g.

For small rodents, the ^{13}C turnover in muscle and liver were measured for *Peromiscus maniculatus* with mass of about 23 g (Miller et al., 2008) and for *Mus musculus* with mass of about 22 g (MacAvoy et al., 2006). *Peromiscus maniculatus* has mass close with *Mus musculus* but a shorter carbon half-time in muscle has been observed contrary to what may be expected. This can probably be explained as a consequence of the higher SusMS and muscle mitochondrial density.

The predicted OBT turnover rates are shorter than the experimental ones for ^{13}C (Miller et al., 2008) in muscle and viscera of *Mus musculus* and can be partially explained by the fact that in the experimental study, the mice were kept captive and not free, as they were considered in the model. An explanation regarding the difference between predicted and experimental half-times for small rodents is related with the improper knowledge of SpMR for animals with body mass lower than 0.2 kg. All allometric relationships used by Wang et al. (2001) and in the present study are deduced for mammals with body mass higher than 0.2 kg. It is generally known that the extrapolation of an allometric relationship for a mass different from the initial mass range considered can be erroneous. SpMR depends also on mitochondrial density in organs. A large difference of mitochondrial density in muscle between athletic and sedentary mammals can be noticed.

An uncertainty factor of 3-4 for half-times was demonstrated for selected mammals and reptiles and it is enough for screening purposes. The study must be expanded to birds and fish before getting a final conclusion and generalisation to a large range of animals (family, species).

In the absence of information for wild animals, data for specific organ metabolic rates for farm and laboratory mammals were used. For wild mammals and birds, the influence of body temperature on metabolic rate was considered and the predicted values of BMR are close to the experimental values (normalised residual less than 40 %).

Any further development of the model and its potential adaptation to a specific site depends on the understanding of the sources of variability for basal (resting, standard) and sustained metabolic rates. The allometric mass relationship of metabolic rate (MR) ($MR=aMass^b$) is not a 'law' but a trend (Hulbert, 2014) and the scaling exponent "b", as well as the intercept "a" show a large variability, especially due to the relative sizes of tissues and organs relevant for the measured activity (allometric models are proposed to represent qualitative trends over orders of magnitude of body mass (Higley and Bytwerk, 2007)).

For radionuclides emitting relatively short range radiations (such as low energy beta radiations) and for organisms above a certain size and complexity, doses to radiosensitive tissues are likely to dictate the resultant radiation effect and consequently, a model based on organs is more appropriate than one based on the whole body as currently used in wildlife models (ICRP, 2008).

Further efforts for model upgrades will be focused on the improving of the data base for organs SpMR of small mammals with mass lower than 0.3 kg and birds and the development of a strategy to assess the consequences of a hypothetical tritium accident for protected species around Cernavoda Nuclear Power Plant (NPP) (Romania). In absence of the specific data for OBT and ^{14}C, the large data base for $^{134,137}Cs$ can be used considering also the animal adaptation to its environment and diet.