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Tritium dynamics in large fish – a model test

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Abstract. Tritium can represent a key radionuclide in the aquatic environment, in some cases, contributing significantly to the doses received by aquatic non-human biota and by humans due to aquatic releases. Recently, the necessity to have a robust assessment of tritium routine and accidental risk emissions for large nuclear installations increased the interest in the topic. In the present study, the recent experiments concerning tritium transfer in adult rainbow trout are described. The updated model concerning the dynamics of tritium transfer in aquatic food chain (AQUATRIT model) developed by the authors is applied and tested for these experimental data. The model predicts the experimental data with a factor of 2 to 3 and the potential improvements of the model are discussed. The present model results emphasize that in the field conditions, the major factors influencing the OBT biological loss rate are the temperature and the prey availability while, the OBT uptake is mainly influenced by the fish growth rates. The main goals of this study are to enhance the robustness of aquatic models for tritium risk assessment and to fulfil a gap for aquatic pathways in environment.

1. INTRODUCTION

Tritium (^{3}H) is a weak beta emitter ($E_{max} = 18$ keV) and is released from some nuclear facilities in relatively large quantities. Tritium is a ubiquitous element because it enters straight into the living organisms as its stable analogue (hydrogen). Tritium can represent a key radionuclide in the aquatic environment, in some cases, contributing significantly to the doses received by aquatic non-human biota and by humans due to aquatic releases. However, the rates of uptake of tritiated water (HTO) and formation of organically-bound tritium (OBT) are currently not very well understood. Tritium enters the food chain in two main chemical forms [1] as does its stable analogue, hydrogen. The *exchangeable* form contains hydrogen molecules bound to elements other than carbon. This constitutes *circa* 70% of body hydrogen predominantly in the form of body water. *Non-exchangeable* hydrogen is found in components such as proteins, lipids and carbohydrates, where it forms a strong covalent bond with carbon. The carbon-hydrogen bond is stable and only broken down by enzyme-mediated reactions. Non-exchangeable hydrogen is generally referred as Organically Bound Hydrogen (OBH, or OBT in the case of ^{3}H).

Apparently, tritium is not an issue of major concern in the aquatic environment because of its rapid dilution in water; but recent events (i.e., the aquatic discharges at Fukushima, Japan and the releases of some very high OBT concentrations in marine biota at Cardiff Bay, UK) increased the interest in the topic, emphasizing the necessity to have a robust assessment of tritium routine and accidental risk emissions for large nuclear installations.

A first attempt to model tritium transfer in aquatic organisms had been done in the past for crayfish [2], but not considering the OBT intake from foodstuff. To improve the understanding of tritium dynamics in aquatic ecosystems, the EMRAS (Environmental Modelling for Radiation Safety) programme coordinated by International Atomic Energy Agency (IAEA) included a Tritium and C-14

Working Group, where many test scenarios were analysed. One such scenario involved the prediction of time-dependent tritium concentrations in freshwater mussels that were subjected to an abrupt change in ambient tritium levels [3]. An updated model concerning the dynamics of tritium transfer in aquatic food chain (AQUATRIT model) was developed and reported recently elsewhere [4]. The experimental data for tritium in large fish are not reported and consequently, AQUATRIT model cannot be tested. The main purpose of the present study is to enhance the robustness of AQUATRIT model for tritium risk assessment, considering the experimental data for large trout.

2. EXPERIMENTAL METHODOLOGY

The experiments were performed at Chalk River Laboratories (Atomic Energy of Canada Limited) in 2009–2010 and cold water species of rainbow trout (*Onchorrynchus mykiss*) were used. The detailed description of the experimental methodology is given elsewhere [5]. The water temperature in tanks was kept constant during the experiment at 13°C after gradual adaptation of fish. Each fish was weighed at the beginning and at the end of each experimental series. The fish were only fed as much food as could be consumed immediately and food consumption was measured daily to determine the feed conversion rate. Experiments were planned for 7, 30, 70 and 140 days (controls and tests). Overall, the amount of food increased during uptake experiments, but significantly decreased during the depuration experiment. The results indicated that growth rates and growing conditions are important for estimating tritium uptake and loss in fish.

For HTO experiment, the average tritium concentration was about: 8100 Bq L^{-1} for the 70 days experiment, 7800 Bq L^{-1} for the 140 days experiment, 8800 Bq L^{-1} for the 30 days, and 8500 Bq L^{-1} for the 7 days experiment, respectively. Water samples from tanks (control and test) were counted by liquid scintillation (Beckman 6500 LSC, Ultima Gold XR) with lower detection limit of 12 Bq L^{-1}. The tissue free water (HTO) of the fish was extracted using a freeze-drying system. For OBT analysis, the tissue remaining after the water extraction was dried at 55°C for 24 hours before proceeding. The dried samples were then homogenized (by scissor and grinder) and mixed with 100 mL of low tritium water to remove the exchangeable OBT from the fish. Samples were refrozen and subjected to a second round of cryogenic distillation under vacuum. The completely rinsed and dried samples were then combusted using a Parr bomb with pressurized oxygen. The combusted water from the fish was diluted to 8 mL with tritium-free water and mixed with 10 mL of Ultima Gold XR. The detection limit for OBT analysis was approximately 55 Bq L^{-1} for fish samples.

3. BRIEF DESCRIPTION OF AQUATRIT MODEL AND ITS ADAPTATION TO TROUT

The general equation of the model for OBT dynamics in consumers, including fish, is:

$$\frac{dC_{org}}{dt} = a\, C_f(t) + b\, C_w(t) - K_{0.5}\, C_{org} \tag{1}$$

where C_{org} is the OBT concentration in fish (Bq kg^{-1} fresh mass (fm)), C_f is the OBT concentration in the food of fish (Bq kg^{-1} fm), a is the transfer coefficient from OBT in the food to OBT in fish (day^{-1}), b is the transfer coefficient from HTO in the water to OBT in fish (day^{-1}), and $K_{0.5}$ is the biological loss rate of OBT from fish (day^{-1}), C_f is the OBT concentration in fish food (Bq kg^{-1} fm).

The OBT biological loss rate, $K_{0.5}$, is given as:

$$K_{0.5} = RGR + R\frac{cal_p}{cal_f} \tag{2}$$

where RGR is the relative growth rate (day^{-1}), R is the respiration rate (g prey fm g^{-1} fish fm day^{-1}), cal_p and cal_f are caloric equivalents of pray (J g^{-1} fm) and fish (J g^{-1} fm), respectively.

Table 1. Model results and comparison with experimental data for HTO uptake case.

exp. duration (days)	fish no. used in each exp.	initial mass (g)	final mass (g)	model OBT conc. (Bq L^{-1})*	exp. OBT conc. (Bq L^{-1})*
7	26	129	151	472.8	475
30	5	448	484	487.5	717
70	4	127	121	941.7	1171
140	4	60	450	1981.6	1534

* Water of combustion.

Table 2. Model results and comparison with experimental data for HTO depuration case: average and standard deviations (sdv).

time (days)	model mass (g)	exp. mass (±sdv) (g)	model OBT conc. (Bq L^{-1})*	exp. OBT conc. (±sdv) (Bq L^{-1})*
140	455.1	448.8 ± 87.9	2045	1534 ± 94.4
170	505.5	484.9 ± 105.8	1513	1214 ± 167.4

* Water of combustion.

In practice, the assessment of $K_{0.5}$ is based on fish bioenergetics models and the detailed description is given in a recent paper [4]. For rainbow trout, the model parameters are considered as those used in the specific bioenergetics models based on experimental verification [6, 7].

4. MODEL RESULTS AND COMPARISION WITH EXPERIMENTAL DATA

4.1 HTO case

Based on the description of the HTO experiment (the input information) [5], the model results are given in Table 1. For each experiment, the initial and final masses of the fish were considered the same as those measured in the experiments and the concentration of HTO in the tank water was considered as an average value of 8400 Bq L^{-1}. Four separate experiments were performed and the initial masses at the beginning and at the end of each experiment are given in Table 1. The model results are close to the experimental data, excepting the day 30, because there is a large individual variability of fish metabolism [8] and quite few fish were used in the experiment. Due to this individual variability, the model can reproduce the data with a factor of 2, in the best case.

For the HTO depuration experiment (Table 2), fish were initially grown for 140 days in contaminated water as in the HTO uptake experiment and the initial fish mass for the depuration experiment was measured. For both model and experiment, the starting value of OBT concentration in fish was considered to be the same as that one for the final OBT concentration in the HTO uptake experiment at day 140. In practice, different fish have been used for the depuration phase, because for any measurement of OBT, the fish must be sacrificed. This implies that the experimental results are statistically biased due to the reduced number of samples in respect to large individual variability. The model results give a theoretical half-life of 144 days for OBT, while the experimental data give an OBT half-life of 185 ± 37 days. Taking into account the individual variability effect, the model reproduces the data with a factor less than 2 and it is close to the experiment, including the experimental errors.

4.2 OBT case

The experimental protocol [5] does not consider the fish tagging, consequently some uncertainties rise for the growth and OBT dynamics. In the experiment, it was assumed that all fish had similar growth

Table 3. Model results and comparison with the experimental data for OBT uptake case: average and standard deviations (sdv).

exp. duration (days)	fish no. used in each exp.	model mass (g)	exp. mass for sacrificed fish (±sdv) (g)	model OBT conc. (Bq L^{-1})*	exp. OBT conc. (±sdv) (Bq L^{-1})*
0	31$	135	128.6 ± 10.8&	0	0
9	3	139.188	132.3 ± 9.1	3823.819	3324 ± 2901
30	7	162.125	290.1 ± 58.6	10657.1	5330 ± 1627
70	3	211.787	254.6 ± 126.6	18458.58	16347 ± 5171
100	2	254.353	237.3 ± 54.9	21750.71	19657 ± 9911
140	7	318.457	310.0 ± 93.3	24352.71	39368 ± 4214

* Water of combustion.
$ The rest of 9 fish were used for the following.
& Initial mass of the 31 fish in tank.

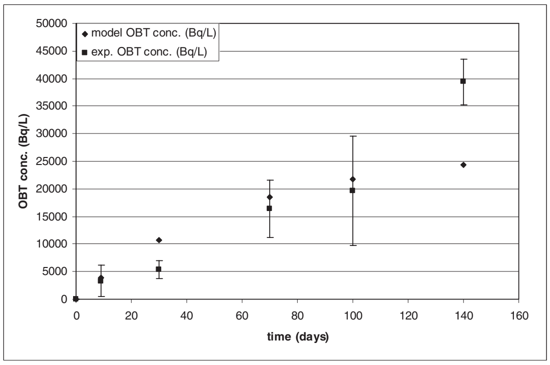

Figure 1. Comparison between model results and experimental data for OBT concentration in fish in the case of OBT uptake.

rates and they were grown under the same condition. The final weights were all measured before sacrifices. The initial values were measured just before the experiment (day 0) and the fish weights are not related to days of exposure group. For example, 31 fish were in each tank and the initial average mass for all 31 fish was 128.6 ± 10.8 g. After 9 days of exposure, three fish were taken from the tank, weighted and sacrificed for the OBT measurements. For the next exposure up to 30 days, the biggest seven fish among the rest of 28 fish, which remained after the 9 days of exposure, were weighted and sacrificed at day 30. In the following exposure duration (up to 70, 100 and 140 days), fish groups were combined from small to large fish randomly.

The model considers that all fish start with the same mass and grow similarly up to a final mass close with the experimental value after 140 days. The model results are compared in Table 3 and Figure 1 with the experimental results, taking into account the effect of individual variability.

In Figure 1, it is observed that the model results are close to the experimental data, excepting the day 30 when the model over predicts the experimental data and the day 140 when the model under predicts the data.

The influence of various growth dynamics on the OBT concentration in fish is given in Figure 2, considering the same final mass of 318 g at day 140, but selecting various initial masses and feeding regimes. The largest OBT concentration is obtained for the normal growth (see the third column in Table 3) and the lowest OBT concentration is obtained in the case of fasting, considering the initial mass of 400 g and the difference between normal feeding regime and fasting is close to a factor of 2.

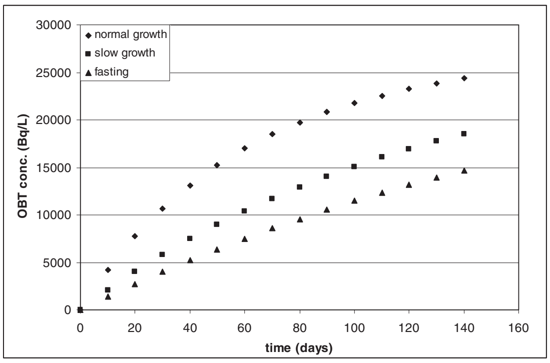

Figure 2. Dependence of OBT concentration dynamics on various feeding regimes for the same final mass.

Table 4. Model results and comparison with the experimental data for OBT depuration case: average and standard deviations (sdv).

time (days)	model mass (g)	exp. mass (±sdv) (g)	model OBT conc. (Bq L^{-1})*	exp. OBT conc. (±sdv) (Bq L^{-1})*
140	318.457	347.6 ± 60.8	24352.71	39368 ± 4212
170	334.096	353.5 ± 62	18751.23	30118 ± 5446

* Water of combustion.

It has to be emphasised that the feeding regime is an important factor and in the experimental conditions there is individual variability in food intake and growth. Further improvements in the experimental methodology must take into account the fish tagging.

For OBT depuration experiment (Table 4), fish were initially grown for 140 days and fed with contaminated OBT food. Seven fish were weighted and were relocated into clean water and fed with clean food. After 30 days, fish were weighted again and sacrificed in order to measure the OBT concentration. For both model and experiment, the starting value of OBT concentration in fish was considered to be the same as that one for the final OBT concentration in the OBT uptake experiment at day 140. The model results give a theoretical half-life of 165 days for OBT, while the experimental data give an OBT half-life of 161.6 ± 46 days. Comparing the experimental OBT half-life after OBT feeding with that one after HTO feeding (185 ± 37 days), it must be noted that they have close values in the limit of the experimental values. In the model, the mass dynamics is slightly different for HTO and OBT depuration, respectively and this explains the small difference between OBT half-lives after HTO and OBT feeding (144 days comparing to 165 days).

5. DISCUSSIONS AND CONCLUSIONS

The updated AQUATRIT model [4] was successfully tested for small fish. In the absence of the experimental data for large fish, it was agreed in 2009 with researchers from Atomic Energy of Canada Limited to use their planned experiments with large trout for a blind test of AQUATRIT model. The main problems were the understanding of experimental conditions and the individual variability influence of metabolic parameters on the both experimental and model results and on their interpretation. The model considered a reference trout, for which the bioenergetics parameters are mainly based on the experimental results for respiration and growth. Consequently, AQUATRIT model was favourably tested for large trout, but the model application to other large fish needs a careful selection of bioenergetics parameters. The bioenergetics model must be first tested for its robustness itself with field and laboratory experiments [9] and then the bioenergetics fish model must be included in the tritium model.

Concerning the biota radioprotection, International Commission for Radiological Protection (ICRP) considers reference animals and the rainbow trout is one of them [10].

The using of the energy metabolism to describe the OBT biological loss rate was proved to be useful for mammals and birds [11], human dosimetry [12] and now, for fish.

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