EMRAS II WG7 – Draft September 2011

Tritium Modelling in Aquatic Systems

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I. Introduction

Tritium (^{3}H) is released from some nuclear facilities in relatively large quantities. It is a ubiquitous isotope because it enters straight into organisms, behaving essentially identically to its stable analogue (hydrogen). Tritium is a key radionuclide in the aquatic environment, in some cases contributing significantly to the doses received by aquatic, non-human biota and by humans.

Models commonly used in tritium dose assessment are steady state specific-activity models based on the assumption of complete isotopic mixing with the stable element, and isotopic equilibrium between all environmental compartments (IAEA, 2010). These models may not be adapted to situations with fluctuating tritium levels in rivers, resulting from discontinuous radioactive discharges or accidental release. To take into account these variations in radioactive discharges, dynamic river, lakes and coastal waters models have been developed (IAEA 2008a, SisBAHIA®, TELEMAC (ref), Mascaret (ref), RIVTOX (Zheleznyak et al., 2000), POSEIDON (Heling et al., 2000)) supplemented by time-dependent food chain models (Ciffroy et al 2005; Galeriu et al 2005; Melintescu and Galeriu, 2011).

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Tritium migration in water bodies is governed by two important processes: (i) the advection of the pollutant by river flow that defines the position of the pollution peak in time and space (advection is fully defined by the river flow velocities), and (ii) the eddy diffusion of the pollutant due to river turbulence, that influences the magnitude of the pollution peak and its spatial spreading (IAEA 2008a).

Tritium interaction with bottom sediments and suspended matter is generally ignored, but some cases were emphasized in case of tritiated water (Turner et al., 2009) or organic matter (Hunt et al., 2010). A minor pathway in terms of dose impact to the population is the tritium transfer between surface water and atmosphere (Marang et al., 2011). For liquid releases, an important pathway is irrigation, but the irrigation effect can be assessed like a precipitation event in terrestrial food chain and it is not included in this document (include a reference to a documents on this pathway). A review of organic tritium in fresh water sediment, animal and plants has been conducted in France (Gontier and Siclet, 2011), it shows that organic tritium from soils (formed over several decades from exposure of vegetation and soil to atmospheric tritium) is the main OBT contributor to the sediments and suspended matter. Recently, the case of dissolved organic tritium (DOT) was treated as a separate pathway of concern for radiopharmaceutical production (Melintescu and Galeriu, 2011).

The importance of tritium transfer in aquatic ecosystems was emphasized in the recent studies in Canada and France (CNSC 2010, ASN 2010) and it is included as a task in the EMRAS II WG7, after a preliminary questionnaire addressed to the participants. In this context, it was agreed to consider only the aquatic tritium food chain transfer.

There are some models of tritium transfer in aquatic organisms developed along the years. The first model of tritium transfer in aquatic organisms was performed for crayfish (Bookhout and White, 1976), but did not consider the OBT intake from foodstuff. In order to update the BURN (Biological Uptake model of RadioNuclides) model (Heling et al., 2002) with a robust tritium sub-model, a new approach was developed within the framework of a contract with KEMA NRG (The Netherlands) (Heling and Galeriu, 2002). Further developments of the model have been reported considering the seasonality and adding a metabolic model for the OBT biological loss rate in fish, as well as a first attempt to consider the Cardiff case (Galeriu et al., 2005). Tritium modelling has been considered in the OURSON (French acronym for Tool for Environmental and Health Risk assessment) model applied to the Loire River (Ciffroy et al., 2006). A simple model was also developed considering a carbon-14 simple model and the ratio between carbon and hydrogen in animals (Sheppard et al., 2006a; Sheppard et al., 2006b). Recently, an updated model of dynamic tritium transfer in the aquatic food chain (AQUATRIT model) was released, using more comprehensive assessments of the aquatic food chain than before, including the benthic flora and fauna, with an explicit application for the Danube ecosystem, as well as an extension to the special case of dissolved organic tritium (DOT) (Melintescu and Galeriu, 2011).

The structure of this document uses the natural sequences in the food chain models from the bottom to the top organisation and includes screening and more complex approaches, if they are available, emphasizing the model performances, if the comparison with the experimental data have been performed. The dynamics of tritiated water in the aquatic organism show a fast equilibration (minutes to hours) with the surrounding water and generally, it is accepted an instant equilibrium:

$$C_{HTO} = C_W * (1 - Dryf) * 0.001 \qquad (1)$$

where: C_{HTO} is the HTO concentration in an aquatic organism (Bq kg^{-1} fresh mass (fm)), C_W is the HTO concentration in water (Bq m^{-3}), 0.001 is the transformation m^3L^{-1}, and Dryf is the dry mass (dm) fraction of an aquatic organism.

For describing the OBT dynamics, the primary producers (*i.e.,* the autotrophs, such as phytoplankton and algae) and the consumers (*i.e.,* the heterotrophs) are treated separately, because the producers convert light and nutrients in organic matter, while the consumers use organic matter from food and add a fraction of organic matter from water through metabolism.

II. Dynamics of organic tritium transfer in producers

OBT dynamics in phytoplankton

In OURSON model (Ciffroy et al., 2006; Siclet F, personal communication, 2009), the basic equation for specific activity of OBT in phytoplankton is the following:

$$\frac{dA_{phyto}^{OBT}(t)}{dt} = k_{photo}\left[DF.A_{water}^{HTO}(t) - A_{phyto}^{OBT}(t)\right] \tag{2}$$

where: A_{phyto}^{OBT} is the specific activity of OBT in phytoplankton (Bq g^{-1} H dm), k_{photo} is the relative gross photosynthetic rate (day $^{-1}$), DF is the isotopic discrimination factor, and A_{water}^{HTO} is the HTO activity in river or sea water (Bq g^{-1} H) = 9.10^{-6} . HTO_{water} (Bq m^{-3}).

Photosynthetic rates k_{photo} vary according to temperature, nutrient availability, solar radiation, etc. Average parameter values can be chosen for each season or more complex models of phytoplankton growth can be used. A default average daily value of 0.5 day^{-1} (averaged over daytime and night time periods, in spring and summer) and a maximum value of 0.1 h^{-1} (representative of the maximum hourly photosynthetic rate) can be used for marine or freshwater phytoplankton. For phytobenthos it is recommended an average value of 0.015 day^{-1} (Riou 1990) and a maximum value of 0.005 h^{-1} , based on measurements of O_2 production by different species of marine benthic algae (Jorgensen 1979). The value of discrimination factor, DF, given in various experiments and reported by Kirchmann et al (1979) is 0.6. and it was emphasized that there is no difference between freshwater and marine environment.

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In AQUATRIT model (Melintescu and Galeriu, 2011), the authors derived and favourably compared the following expression with experimental data (Heling and Galeriu, 2002; Galeriu et al., 2005):

$$\frac{dC_{o,phpl}}{dt} = 0.4 \cdot \mu \cdot Dryf \cdot 0.001 \cdot C_W - \mu \cdot C_{o,phpl} \tag{3}$$

where: $C_{o,phpl}$ is the OBT concentration in phytoplankton (Bq kg^{-1} fm), and μ is the phytoplankton growth rate (day^{-1}).

The phytoplankton growth rate depends on the nutrients in water, light, and water temperature. The details are given elsewhere (Melintescu and Galeriu, 2011).

OBT dynamics in macrophyte

In OURSON model, the same equation as for phytoplankton (Eq. 2), it is used for macrophyte:

$$\frac{dA_{plant}^{OBT}(t)}{dt} = k_{photo} \cdot \left[DF.A_{tissue-water}^{HTO}(t) - A_{plant}^{OBT}(t)\right] \tag{4}$$

If part of the plant body is at or above the water surface, HTO will equilibrate between water and atmosphere and the specific activity of HTO in the plant tissue water can be considered to be equal to the average between HTO in river water and HTO in air moisture.

For the assessment of the OBT concentration in macrophytes following an accidental contamination, AQUATRIT model uses the same equation as for phytoplankton (Eq. 3), but a specific growth rate. The growth processes of macrophytes are described in the literature (Herb and Stefan, 2006; Hakanson and Boulion, 2002). The growth rate depends on the species, temperature, water turbulence, water depth where the plants grow, and water surface irradiance; it can vary widely, depending on local conditions. For the model application in a specific case, the above general theory (Herb and Stefan, 2006; Hakanson and Boulion, 2002) is used for local conditions. In AQUATRIT model, applied for Danube ecosystem, benthic algae are considered to have a maximum growth rate of 0.01 day^{-1}, depending on water temperature, and daily average irradiance, given by:

$$\mu_{ba} = 0.01 * 1.07^{(T-8)} * \mathrm{mod}\,light^{0.31} \tag{5}$$

where: μ_{ba} is the growth rate of benthic algae (day^{-1}), T the average water temperature (ºC), and mod*light* the moderator of seasonal irradiance variability, considered the same one as for phytoplankton.

The approach considered in AQUATRIT model is conservative, ignoring the discrimination factor (*i.e.* the ratio between tritium and hydrogen, T/H) of about 0.6, used in the recent recommendations (IAEA 2010).

The mass fraction of dry matter in benthic algae has a mean value of 0.08, but some default values for water content of various aquatic organisms are given in Table 87 in TRS 472 (IAEA 2010). The growth rate used in the description of the Danube ecosystem is not generally valid, and variations by a factor of 3 in this parameter are expected for other local conditions.

III. Dynamics of organic tritium transfer in consumers

In OURSON model, the following description refers to fish but it can be applied to molluscs and crustaceans, as well. The model assumes that the animal organic biomass can be represented by a single compartment and the OBT turnover in biotic compartments has the same characteristics as carbon turnover. The model takes also into account the mass balance of OBT and the evolution of individual fish biomass which is equal to the difference between the gain through ingestion and the loss through respiration. After preliminary calculations given elsewhere (Sheppard et al., 2006a), it stated the basic equation for the specific activity of OBT in fish:

$$\frac{dA_{fish}^{OBT}(t)}{dt} = k_{ing} \cdot \left[\frac{H_{diet}}{H_{fish}} . A_{diet}^{OBT}(t) - A_{fish}^{OBT}(t) \right] \tag{6}$$

with

$$k_{ing} = I.D$$

where: A_{fish}^{OBT} is the OBT specific activity in fish (Bq g^{-1} H dm), k_{ing} is the relative ingestion rate (day $^{-1}$), H_{diet} is the mass ratio between hydrogen and carbon in diet (g H g^{-1} C), H_{fish} is the mass ratio between hydrogen and carbon in fish dry matter (g H g^{-1} C), A_{diet}^{OBT} is the specific activity of OBT in diet (Bq g^{-1} H dm), I is the relative food intake rate (day $^{-1}$), and D is the feed digestibility.

The turnover rate of OBT finally depends on two metabolic parameters, the relative food intake rate of fish *I* (kg of ingested C per kg of C in fish biomass) and the feed digestibility. The average values of the relative ingestion rate, k_{ing}, are given in Table 1 and the ratio between hydrogen and carbon in diet, H/C, (g H g^{-1} C) is given in Table 2.

Comment [AM4]: I depends on fish mass, also. Please, indicate some ranges for I and D, in order to apply the model.

I and D are not available for fish in natural environment, the parameter king (IxD) which is the sum of the metabolic rate and the growth rate is easier to estimate.

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Table 1. Average values of relative ingestion rate for aquatic fauna

Animal type	k_{ing} (day^{-1})	Reference
Fish	0.001	Sheppard et al. (2006b)
Mussel	0.02	IAEA (2008b)
Shrimp (aquaculture Madagascar)	0.1	Franco et al. (2006)

Table 2. Empirical hydrogen to carbon ratios in various biotas obtained from environmental monitoring of French NPP

Type of biota	H/C
Phytoplancton	0.16[1]
Macrophytes	0.14
Fish	0.15
Mussel	0.17
Shrimp	0.15

[1] theoretical ratio of photosynthesis

In OURSON model, the equations are based the specific activity approach, but in practice, the concentrations in fresh mass are needed. To cope with this need, OURSON uses the following conversion equations for HTO and OBT, respectively:

$$\begin{aligned} C_{fw}^{HTO} &= WC \cdot C^{HTO} \\ C_{fw}^{OBT} &= (1-WC) \cdot WEQ \cdot C^{OBT} \end{aligned} \qquad (7)$$

where: C_{fw}^{HTO} is the HTO concentration in biota (Bq kg^{-1} fw), C_{fw}^{OBT} is the OBT concentration in biota (Bq kg^{-1} fw), *WC* is the fractional water content of the organism (kg water kg^{-1} fw), *WEQ* is the water equivalent factor of the organism (i.e. volume of water obtained by combustion of dry tissue) (L kg^{-1} dm), $C^{HTO} = 111 * A^{HTO}$ is the tritium concentration in tissue free water (Bq L^{-1}), $C^{OBT} = 111 * A^{OBT}$ is the tritium concentration in combustion water (Bq L^{-1}), A^{HTO}, A^{OBT} are the tissue HTO and OBT specific activities, respectively (Bq g^{-1} H)

Values of WC for various aquatic organisms are available in Table 87 of TRS 472 (IAEA 2010). Values of WEQ are given in Table 3.

Table 3. Water equivalent factors (WEQ) for various aquatic organisms

Organism	Water equivalent factor (g water g^{-1} DW)	Reference
Marine algae	0.50	EDF*
Marine fish	0.65	EDF*
Molluscs (soft part)	0.60	EDF*
Crustaceans (soft part)	0.60	EDF*
Freshwater fish	0.65	IAEA (2010)

*- empirical values from radioecological monitoring of NPP

In AQUATRIT model, for all the other aquatic organisms (zooplankton, crustaceans, molluscs, and fish), the OBT concentration dynamics, including the specific hydrogen (tritium) metabolism, is well described in a previous paper (Galeriu et al., 2005). The general equation for OBT dynamics in consumers is:

$$\frac{dC_{org,x}}{dt} = a_x\, C_{f,x}(t) + b_x\, C_w(t) - K_{0.5,x}\, C_{org,x} \qquad (8)$$

where $C_{org.x}$ is the OBT concentration in the animal, x (Bq kg^{-1}fm), $C_{f,,x}$ is the OBT concentration in the food of animal, x (Bq kg^{-1}fm), a_x the transfer coefficient from OBT in the food to OBT in the animal, x (day^{-1}), b_x the transfer coefficient from HTO in the water to OBT in the animal, x (day^{-1}), and $K_{0.5,x}$ the biological loss rate of OBT from animal, x (day^{-1}).

For a proper mass balance, it is necessary to introduce the following relationship (Galeriu et al., 2005):

$$C_f = \sum_{i=1}^{n} C_{prey,i}\, P_{prey,i}\, \frac{OBH_{pred}}{OBH_{prey,i}} \qquad (9)$$

where C_f is the OBT concentration in animal's food (Bq kg^{-1}fm), $C_{prey,i}$ the OBT concentration in prey, i (Bq kg^{-1}fm); $P_{prey,i}$ the preference for prey, i, and OBH_x the organically bound hydrogen (OBH) content in organism, x (prey or predator) (g OBH kg^{-1} fm).

In the absence of the relevant data, the ratio of OBH in predator and prey can be assessed from the dry matter ratio, with a moderate loss of accuracy.

Equations 7 and 8 refer to a model with a single OBT compartment with more than one source of OBT production: from HTO in water or OBT in food. When HTO dominates as the primary source, the specific activity approach can be used. The specific activity (SA) of tritium is defined as the ratio between the tritium activity and the mass of hydrogen in a specific form. The specific activity ratio (SAR) is the ratio between the SA of OBT in the animal and the SA of HTO in water. Based on a literature review (Heling and Galeriu, 2002; Galeriu et al., 2005), the values for SAR in different aquatic organisms when the source is HTO is given in Table 4. not consistent with IAEA 2010 which recommends an average value of 0.66 (page 138 in TRS 472)

Comment [MSOffice6]: I don't agree with this. We refer to SAR and TRS 472, (p.138) refers to partition fractions. We distinguish between OBT in fish coming from water metabolism and coming from OBT in food. TRS doesn't do this. If you take into consideration this, the values in both refs. are comparable.

Table 4. Specific activity ratio **(**SAR) and standard deviations (sd) for aquatic organisms when the source is HTO

Aquatic organisms	**SAR (HTO source) ± sd**
Zooplankton	0.4±0.1
Molluscs	0.3±0.05
Crustaceans	0.25±0.05
Planktivorous fish	0.25±0.05
Piscivorous fish	0.25±0.05

Using the specific activity approach and the equilibrium conditions, the transfer coefficients in Eq. 8 are now defined as:

$$a_x = (1 - SAR_x) * K_{0.5,x}$$

$$b_x = SAR_x * K_{0.5,x} * \frac{SA_{pred}}{111} \tag{10}$$

where: SAR_x is the specific activity ratio in animal, x, SA_{pred} the specific activity of bound hydrogen (BH) in the predator (kg BH kg^{-1} fm), and 111 the mass of free hydrogen (kg) in 1 m^3 of water.

With the exception of fish fat, SA_{pred} is about 0.06*$Dryf_{pred}$, depending on the dry matter fraction of the predator. For fish fat, a value of 0.08*$Dryf_{pred}$ is recommended for SA_{pred}.

OBT dynamics in zooplankton

In AQUATRIT model, the OBT biological loss rate, $K_{0.5}$, for zooplankton depends on its growth rate and temperature (Ray et al., 2001). At a reference temperature of 20 ºC and considering the zooplankton volume, the OBT biological loss rate is given by:

$$K_{0.5\_o} = (0.715 - 0.13 * \log(V)) + (0.033 - 0.008 * \log(V)) \tag{11}$$

where $K_{0.5\_o}$ is OBT biological loss rate at the optimal reference temperature of 20 ºC (d^{-1}), and V the zooplankton volume (μm^3).

The dry matter fraction of zooplankton varies between 0.07 and 0.2; in AQUATRIT model, a value of 0.12 is used as a default value. All the details are given elsewhere (Melintescu and Galeriu, 2011).

OBT dynamics in zoo benthos

In AQUATRIT model, the benthic fish consume macrointervertebrates and especially, aquatic insect larvae of the Order Diptera. The most widespread ones are those from the Chironomidae (or chironomid) Family, which has two to six life cycles per year. Generally, chironomid larvae are assumed to have a growth rate of 0.05 day^{-1} and a respiration rate of 0.01 day^{-1} (Heling 1995). Consequently, the OBT biological loss rate for chironomid larvae, $K_{0.5}$, is 0.06 day^{-1} (Heling 1995). A higher value ($K_{0.5}$ = 0.2 day^{-1}) is used in the CASTEAUR (French acronym for Simplified CAlculation of radioactive nuclides Transfer in Receiving WATERways) model (Beaugelin-Seiller et al., 2002). In AQUATRIT model, an average value, $K_{0.5}$ = 0.1 day^{-1} is used. All the previous values for $K_{0.5}$ correspond to an average water temperature of 12 ºC. In the absence of relevant data, for other water temperatures, the temperature correction functions were considered as those for molluscs and crustaceans.

Small molluscs and crustaceans have a very large variability, and the calculations of their OBT biological loss rates must be adapted to different cases. For molluscs, a literature review (Heling and Galeriu, 2002) gives a $K_{0.5}$ of 0.02 day^{-1} for a body mass of 1 g fm, but a $K_{0.5}$ of 0.005 day^{-1} is used for 30 g of soft tissue. For crustaceans, the same review (Heling and Galeriu, 2002) cites an average value of 0.007 day^{-1} for $K_{0.5}$. By comparison, for molluscs, a value of 0.017 day^{-1} for $K_{0.5}$ is given in the literature (Heling 1995). Based on experimental data for the growth rate and the energy content of *Mytilus edulis* soft tissue (2,386 J per g wet tissue), the following relationship can be derived (Sukhotin et al., 2002):

$$K_{0.5\_o} = 0.024 * W^{-0.246} \tag{12}$$

where W is the wet mass of mussel soft tissue (g fm)

Recent experiments concerning OBT dynamics for *Elliptio complanata*, with a total mass of 90 g (40 g wet mass), give a value of 0.02 day^{-1} for $K_{0.5}$ (IAEA 2008b; Yankovich et al., 2011), a value which is a few times higher than that for *Mytilus edulis* (Sukhotin et al., 2002).

For the food chain modelling, molluscs and crustaceans are of interest, since they are consumed by humans and various species of zoo-benthos are consumed by fish. The model has two separate compartments. For human consumption, mussels and crabs of large body mass (about 20 g fm for both mussels crabs) are included and the model parameters are adapted to approximate this mixture. By default, a biological loss rate of 0.007 day^{-1} is assumed for OBT, but model users must adapt this value to their specific cases.

The temperature dependence of the OBT biological loss rate for molluscs and crustaceans is considered, based on experimental data for a *Tridacna* species (Hean and

Cacho, 2003), without any guarantee that it is correct for the specific applications (*i.e.*, the cases considered for AQUATRIT model).

There is large variability in the influence of body mass and temperature on aquatic invertebrate respiration (Brey 2010), and in specific cases, the literature must be consulted for improved parameters.

OBT dynamics in fish

There are very few experimental data for OBT biological loss rates in fish. In the absence of experimental data, models based on bioenergetics are used here, as it has been experimentally demonstrated that the mass dependence of basal metabolic rate of fish is a combination between the tissue-specific respiration rate and the relative size of different tissues (Oikawa and Itazawa, 2003). The same approach as for mammals (*i.e.,* the energy metabolism approach) (Galeriu et al., 2009) can also be considered for aquatic fauna for tritium transfer.

Bioenergetics involves the investigation of energy expenditure, losses, gains and efficiencies of transformations in the body. The basic equation for bioenergetics models (BEMs) of fish growth is as follows (Hanson et al., 1997):

$$\frac{1}{W}\frac{dW}{dt} = [C-(R+S+F+E+P)]\frac{cal_p}{cal_f} \tag{13}$$

where W is the fish mass (g fm), t the time (day), C the consumption (g prey g^{-1} fish day^{-1}), R the respiration or losses through metabolism (g prey g^{-1} fish day^{-1}), S the specific dynamic action or losses because of energy costs of digesting food (g prey g^{-1} fish day^{-1}), F the egestion or losses through faeces (g prey g^{-1} fish day^{-1}), E the excretion or losses of nitrogenous wastes (g prey g^{-1} fish day^{-1}), P the egg production or losses through reproduction (g prey g^{-1} fish day^{-1}), and cal_p, cal_f are caloric equivalents of pray (J g^{-1}) and fish (J g^{-1}), respectively.

The equation for consumption is:

$$C = C_{\max} * p * f_c(T) \tag{14}$$

where C is the consumption (g prey g^{-1} fish day^{-1}), C_{max} the allometric equation for maximum specific consumption rate (g prey g^{-1} fish day^{-1}), with $C_{max} = aW^b$ with a, b are allometric coefficients for fish, p the proportion of maximum consumption, and $f_c(T)$ a temperature-dependent function.

Respiration is measured as oxygen consumption and it is converted to consumed prey, by knowing the energy equivalent of oxygen (13,560 J g^{-1} O_2) and the prey energy density. Respiration depends on temperature, fish mass (allometric function) and activity:

$$R = a_r W^{br} f_r(T) ACT * conv \tag{15}$$

where R is the respiration (g prey g^{-1} fish day^{-1}), a_r, b_r are allometric coefficients (a_r is usually given in units of O_2 consumption per g fish and unit time), $f_r(T)$ the temperature function of respiration, ACT an activity multiplier depending on fish average swimming speed, and conv is the oxygen consumption converted to consumed prey (13,560 J g^{-1} O_2 cal_p^{-1}).

Note that all the units in Eqs. 13-15, are reported on a fm basis.

In many applications, the specific dynamic action (S), the egestion (F), and the excretion (E) depend on consumption, as an overall fraction (ε), and the relative growth rate (RGR) is given by Eq. 16:

$$RGR = \frac{1}{W}\frac{dW}{dt} = [(1-\varepsilon)C - R]\frac{cal_p}{cal} \tag{16}$$

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

$$K_{0.5} = RGR + R\frac{cal_p}{cal_f} \tag{17}$$

In Eq. 17, the effect of growth dilution (RGR) and the metabolic (respiration) rate must be noted.

At maintenance (RGR=0), the OBT biological loss rate is given only by respiration. The development and application of BEMs increased substantially in the last decade. BEMs are appealing because they are based on balanced energy-fate equations that have been thought to promote reasonable predictive behaviour. However, most BEMs have not been well evaluated over the ranges of conditions to which they have been applied. Results indicate that many BEMs are substantially inaccurate when predicting fish growth with higher feeding rates or estimating consumption with higher growth rates, even when higher consumption levels or growth episodes are of short duration (Bajer et al., 2004). Further work is needed to evaluate temperature, sub-maintenance-feeding, and prey-type effects on the performance of BEMs, as well as possible influences of swimming activity level (*i.e.*, ACT in Eq. 14). In a recent review (Chipps and Wahl, 2008), BEMs have been analyzed in relation with field and laboratory experimental data. Field tests of bioenergetics models have generally revealed poor fits between model predictions and field estimates; however a reasonable agreement (15 %) was obtained between model and field values for lake trout (*Salvelinus namaycush*), largemouth bass (*Micropterus salmoides*), and sockeye salmon (*Oncorhynchus nerka*) (Chipps and Wahl, 2008). Laboratory tests also show poor performance (Bajer et al., 2004). Disagreement between BEMs and laboratory data are largest when attempting to account for a range of temperatures and variable ration levels on model estimates. Subtle physiological adaptations of fish species to local environment can also have an important influence on the accuracy of BEMs predictions.

IV. Dissolved organic tritium (DOT)

The models previously described (OURSON and AQUATRIT) are based on the assumption that the OBT specific activity in fish is directly linked with the HTO in water or the OBT in fish food. This is fully valid if the water contamination is due only to an initial HTO source. Taking into account this supposition, the concentration factor (CF) in fish must be less than or equal to 1. Classically, the concentration factor has been defined as the ratio between the concentration per unit mass of biota at equilibrium and the dissolved concentration per unit volume in ambient water.

For the marine environment at Cardiff, UK, CFs for tritium in biota have been investigated (McCubbin et al., 2001; Williams et al., 2001). For flounder (*Platichthys flesus*) and mussels (*Mytilus edulis*), CFs of up to 4×10^3 (fresh mass equivalent) were reported. The significant increase in CF compared with unity has been attributed to uptake of tritium in organically bound forms, due to the existence of organic species of tritium in a mixture of compounds in the authorized releases of wastes to the Bristol Channel from the Nycomed-Amersham (now GE Healthcare) radiopharmaceutical plant at Whitchurch, Cardiff, UK. A review of past monitoring results was recently published (Hunt et al., 2010) and difficulties with analytical methods regarding OBT have been pointed out. The extremely large CFs cannot be explained by analytical errors and many hypotheses have been advanced. These include concentration of organic tritium by bacteria, and subsequent transfer in the food chain; ingestion of contaminated sediment; ingestion of contaminated prey; and direct uptake of DOT from the sea water. It was suggested that bioaccumulation occurs via a pathway for the conversion of the tritium-labelled organic compounds into particulate matter (via bacterial uptake / physico-chemical sorption) and the subsequent transfer to the food chain (McCubbin et al., 2001). Comparison of monitoring data for sediment and suspended matter with data on tritium in benthic fauna shows that the ingestion of sediment or particulate matter is not a reasonable explanation, since the OBT concentration in benthic fauna is much higher than the OBT concentration in both sediments and suspended matter. Is the specific activity of OBT in benthic fauna, in sediment and suspended matter available?

Comment [MSOffice7]: Specific activity referring to what kind of source??? Please, be more specific.
And Yes, there are, but it depends on the source. SA of OBT in this case refers to DOT source. You may contact by yourself FSA, UK to provide their private reports for this case.

Assuming that molecules of DOT are highly bio available, a conservative approach considers that the dissolved organic compounds are the only carbon source for aquatic plants and animals. Then, OURSON equation (6) for the transfer of organic tritium to consumers can be used by replacing the specific activity in the diet with the specific activity in DOT. Similarly, In OURSON, the equations (2) and (4) for the OBT dynamics in phytoplankton and macrophytes, respectively can be used by replacing DF.A^{HTO} with the specific activity in DOT. The turn-over rate depends on the relative carbon intake rate. Thus, the kinetic parameters previously described, k_{photo} and k_{ing} can also be applied to plant and animal uptake of dissolved organic molecules. The specific activity of organic tritium in dissolved organic matter A_{DOM}^{OBT} (in Bq/g H) is expressed as:

$$A_{DOM}^{OBT} = \frac{C_{water}^{OBT}}{DOC.H_{DOM}} \qquad (18)$$

where: C_{water}^{OBT} is the organic tritium activity in filtered river or sea water (Bq L^{-1}), *DOC* is the dissolved organic carbon concentration in river or sea water (g L^{-1}), H_{DOM} is the hydrogen to carbon mass ratio in dissolved organic matter (theoretical ratio of 0.166 corresponding to 2 atoms of hydrogen for 1 atom of carbon) (g H g^{-1} C).

The dissolved organic carbon, DOC, for various aquatic ecosystems is given in Table 5.

Then, activity in plant and animal can be calculated with the following equations, assuming DOM is the only carbon source for the plant or animal (conservative assumption):

$$\frac{dA_{plant}^{OBT}(t)}{dt} = k_{photo} \cdot \left[\frac{H_{DOM}}{H_{plant}} . A_{DOM}^{OBT}(t) - A_{plant}^{OBT}(t) \right] \tag{19}$$

$$\frac{dA_{animal}^{OBT}(t)}{dt} = k_{ing} \cdot \left[\frac{H_{DOM}}{H_{animal}} . A_{DOM}^{OBT}(t) - A_{animal}^{OBT}(t) \right] \tag{20}$$

where: H_{DOM} is the mass ratio between hydrogen and carbon in DOM (theoretical ratio of 0.166 corresponding to 2 atoms of hydrogen for 1 atom of carbon) (g H g^{-1} C), H_{plant} is the mass ratio between H and C in the aquatic plant (g H g^{-1} C), H_{animal} is the mass ratio between H and C in the aquatic animal (g H g^{-1} C), A_{DOM}^{OBT} is the specific activity of organic tritium in dissolved organic matter (Bq g^{-1}H), A_{plant}^{OBT} is the specific activity of organic tritium in the aquatic plant (Bq g^{-1}H), and A_{animal}^{OBT} is the specific activity of organic tritium in the aquatic animal (Bq g^{-1}H).

Table 5. Dissolved organic matter parameters

Water body	DOC (mg C L^{-1})	Reference
Loire River	3	Abr 02
Loire Estuary	9	Abr 02
English Channel	2	Abr 02
North Pacific	0.9	Peltzer and Hayward, 1996

Comment [AM8]: Please, give this reference. I will send it later, I don't have it on my computer

Comment [AM9]: Please, give this reference complete: title, journal, no, vol., year, etc. I will send it later

In AQUATRIT model, the direct uptake of DOT can be introduced in the dynamic equation for phytoplankton (Eq. 3) and consumers (Eq. 8), respectively:

$$\frac{dC_{o,phpl}}{dt} = 0.4 \cdot \mu \cdot Dryf \cdot 0.001 \cdot C_W + V_{DOT} * C_{DOT} - \mu \cdot C_{o,phpl} \tag{21}$$

$$\frac{dC_{org,x}}{dt} = a_x \, C_{f,x}(t) + b_x \, C_w(t) + V_{DOT} \cdot C_{DOT} - K_{0.5,x} C_{org,x} \tag{22}$$

where C_W is the HTO concentration in water (Bq m^{-3}), C_{DOT} the DOT concentration (Bq L^{-1}), and V_{DOT} the uptake rate of DOT (L kg $^{-1}$fm day^{-1}) and it is obtained from a simplified form of Michaelis-Menten equation (full details are given elsewhere (Strack et al., 1980; Melintescu and Galeriu, 2011).

V. Examples of AQUATRIT model application for a typical fish

Comment [AM10]: Francoise, if you applied and tested the OURSON to different types of fish, please provide examples, in order to be embed into the draft.

We have to discuss if it is necessary to give examples, this has been done already in the previous EMRAS TECDOC.

This is the dynamic accidental case, not like the previous case. And in that previous case, you tested the model to molluscs, not to fish. And to test the model with the data is the MOST IMPORTANT thing. We may develop as many models we like, but without testing them with a data, they mean nothing.

In case you don't test the model with fish data is ok.

In this example, we choose the rainbow trout (*Oncorhynchus mykiss*), because it is a representative fish in many countries and it is also considered as a representative fish by ICRP (ICRP 2008).

For rainbow trout (*Oncorhynchus mykiss*), the OBT dynamics was studied for juveniles (Rodgers 1986), and adults (Kim SB, personal communication, 2010). Juvenile rainbow trout were kept in tritiated water and/or received a diet labelled with tritiated amino acids at a constant temperature of 15 °C. The average mass of fish increased from 7.0 ± 0.2 g fm up to 15.7 ± 0.6 g fm during the course of the 10 week experiment (with 56 days for tritium uptake). Based on the experimental data during exposure to a tritiated diet, the OBT rate constant was 0.0218 ± 0.002 day^{-1}, while in the next two weeks after exposure, the estimated value was 0.0308 ± .0031 day^{-1}. For the juvenile rainbow trout model in AQUATOX (Park and Clough, 2009), the OBT rate constant for the experimental condition, such as mass range and water temperature, considered was close to 0.03 day^{-1}. More recently, an updated model for rainbow trout was successfully used (Tyler and Bolduc, 2008). When this last model was applied for OBT dynamics, the rate constant was 0.037 day^{-1}. These results must be considered with caution, however, because under laboratory conditions, the fish activity is lower than under field conditions and the models use a mixture of parameters that are not fully coherent.

The bound hydrogen (BH) content and energy density (ED) in fish and its prey can be assessed knowing the composition of fish and its prey regarding carbohydrates, proteins and lipids. The values of BH and ED per kg of carbohydrates, proteins and lipids are found in literature (Diabate and Strack, 1993; Murray and Burt, 2001). The model needs the OBT biological loss rate (defined by eq. 16) which is obtained using fish bioenergetics models tested with laboratory and field data on fish growth. The fish bioenergetics theory is well established and the software and the default data base are available (Hanson et al., 1997). The most appropriate parameters for the fish of interest must be found in the recent literature and the end users must be careful in choosing the young and adult fish cases. In case of rainbow trout, for the youngest fish (mass less than 50 g) the parameters are given elsewhere (Tyler and Bolduc, 2008). For the adult fish, different parameters are recommended (Railsback and Rose, 1999; Rand et al., 1993). The referenced papers (Railsback and Rose, 1999; Rand et al., 1993) use the experimental data on fish respiration in standard and active state, as well as data on fish growth for controlled nutrition. These requirements are essential for tritium models applications. For the laboratory experiments at Chalk River Laboratory (AECL, Canada), the model was blind tested and after it was compared to the final results, the predicted to observed ratios were less than a factor of 2 (Figure 1) and the discrepancies between the model and the data can be partially explained by the unknown details on fish mass dynamics in the experiment (Melintescu et al. 2011).

For the model application in realistic field conditions, other important information is needed, considering the prey composition and energy density, as well as prey availability (Megrey et al., 2007). The seasonal variability of prey (composition and density) influences the fish growth and considering the seasonal water temperature variability also, the OBT concentration in fish may largely vary.

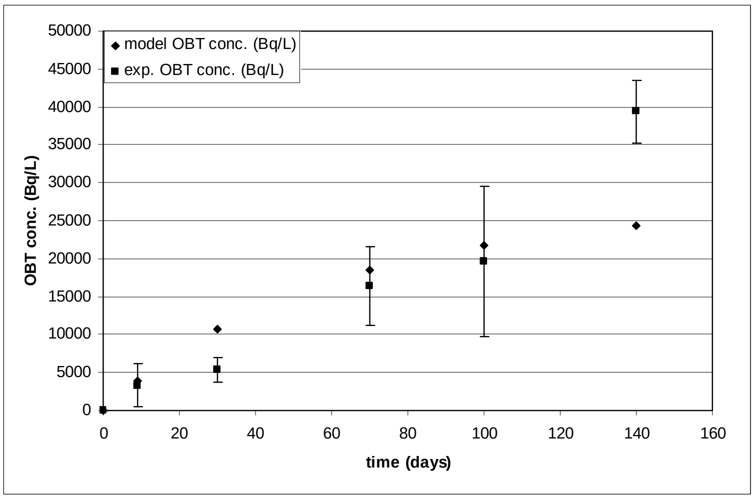

Figure 1. Comparison between model results and experimental data for OBT concentration in fish in the case of OBT uptake

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