

UPDATED AQUATRIT AS FOR USERS

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 Tritium (³H) - not a problem of major concern in aquatic environment (apparently) → dilution in water.

Recently, two events increased the interest in the topic:

1. The necessity to have a robust assessment of tritium routine and accidental risk emissions for large nuclear installations;

2. The releases of some very high OBT concentrations in marine biota at Cardiff Bay (UK), unexpectedly for the pre-supposition that it is not bioaccumulation of tritium in aquatic biota (Williams et al., 2001).

Modelling attempts

- ³H transfer in aquatic environment started in 1970s in USA with a series of experiments and few modelling trials and the results have been summarized in a dedicated paper (Blaylock et al., 1986) → The conclusion was that the dose coming from the ingestion of aquatic foodstuff is lower than the dose coming from the intake of HTO in water.
- A first attempt to model ³H transfer in aquatic organisms had been done in the past for crayfish (Bookhout and White, 1976), but not considering 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 had been developed in a frame of a contract with KEMA NRG (The Netherlands) (Heling and Galeriu, 2002).
- Further development of the model have been reported, considering the seasonality and adding a metabolic model for OBT biological loss rate in fish, as well as a first attempt to consider the Cardiff case (Galeriu et al., 2005).
- More recently, tritium modelling has been considered in OURSON (French acronym for Tool for Environmental and Health Risk assessment) model applied to Loire River (Ciffroy et al., 2006), but the fish sub-model is not proper defined (Siclet F Personal communication 2009).

Updated AQUATRIT model

- Dynamic model for predicting ³H transfer in aquatic food chain;
- More coherent assessments for the aquatic food chain, including the benthic flora and fauna;
- Explicit application for the Danube ecosystem;
- Model extension to the specific case of dissolved organic tritium (DOT)

Model description

T in the body water of the animal is considered in equilibrium with the HTO in the aquatic environment, and is expressed by the following simple relationship (Galeriu et al., 2005):

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

- C_{HTO} HTO concentration in aquatic organism (Bq kg⁻¹ fw);
- C_W HTO concentration in water (Bq m⁻³);
- 0.001 the transformation m³L⁻¹;
- Dryf the dry mass (dm) fraction of aquatic organism

For describing the OBT dynamics, the primary producers (*i.e* the autotrophs as phytoplankton and algae) and the consumers (*i.e* the heterotrophs) are treated separately.

OBT dynamics in phytoplankton

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

C_{o,phpl} - OBT concentration in phytoplankton (Bq kg⁻¹ fw);

- phytoplankton growth rate (day⁻¹) depends on: nutrients in water, light, water temperature
- the updated present model considers an optimal growth rate of phytoplankton, $\mu_o = 0.5 \text{ day}^{-1}$

$$\mu = \mu_0 * \mod light * \mod temp$$
 modlight – seasonal light moderator;
modtemp – seasonal temperature moderator

• The relative seasonal variability of light is chosen as following:

 $mod light = min + (1 - min) * sin(\pi * \frac{julianday}{365})$ min – the ratio between minimum irradiance (winter time) and maximum irradiance (summer time), chosen as min=0.3, based on local conditions

• The temperature moderator (using local data or generic relationships for rivers which depend on latitude) is:

 $mod temp = 1.065^{(T-20)}$ T – water temperature (°C)

OBT dynamics in macrophyte

- For the assessment of the OBT concentration in macrophytes, we use the same equation as for phytoplankton, but using a specific growth rate, μ.
- µ depends on: species, temperature, water turbulence, water depth where the plants grow, and water surface irradiance and can largely vary, depending on local conditions.
- For Danube ecosystem, the turbulence is moderate to high and water depth varies pretty much.
- The algae grow more towards the shore and there are considered average characteristics in order to derive a simplified model, where both the temperature and the irradiance are generic parameters.
- In the present model, we consider benthic algae with a maximum growth rate of 0.01 day⁻¹, depending on water temperature, and daily average irradiance, and given by:

$$\mu_{ba} = 0.01 * 1.07^{(T-8)} * \text{mod } light^{0.31}$$

OBT dynamics in consumers

- 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:

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

 $\begin{array}{l} \mathsf{C}_{\text{org},x} \\ \mathsf{C}_{\text{f},x} \\ \mathsf{a}_{x} \end{array} \stackrel{\text{-} \mathsf{OBT}}{\text{concentration in the animal x (Bq kg^{-1}fw);} \\ \mathsf{a}_{x} \\ \mathsf{a}_{x} \end{array} \stackrel{\text{-} \mathsf{OBT}}{\text{concentration in the food of animal x (Bq kg^{-1}fw);} \\ \mathsf{b}_{x} \\ \mathsf{c}_{\text{transfer coefficient from OBT in the food to OBT in the animal x (day^{-1});} \\ \mathsf{b}_{x} \\ \mathsf{b}_{x} \\ \mathsf{c}_{0.5,x} \end{array} \stackrel{\text{-} \text{biological loss rate of OBT from animal x (day^{-1})} \end{array}$

• For a proper mass balance, we have:

$$C_{f} = \sum_{i=1}^{n} C_{prey,i} P_{prey,i} \frac{OBH_{pred}}{OBH_{prey,i}}$$

 C_f - OBT concentration in animal's food (Bq kg⁻¹fw);

C_{prey,i} - OBT concentration in prey,i (Bq kg⁻¹fw);

- $P_{prey,i}$ the preference for prey,i;
- OBH_x organically bound hydrogen (OBH) content in organism x (prey or predator) (g OBH kg⁻¹ fw)

- The previous equations refer to a model with a single OBT compartment and with different sources of OBT productions: from HTO in water or OBT in food.
- It is considered that having only HTO as a primary source, the SA approach can be used.

SAR and standard deviations (sdv) for aquatic organism when the source is HTO

Aquatic organisms	SAR (HTO source) ± sdv		
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		

SA=T activity / mass of H corresponding to the specific form

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SAR = SA of OBT in the animal / SA of HTO in water
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Using the specific activity approach and the equilibrium conditions, the transfer coefficients are now defined as:

$$a_{x} = (1 - SAR_{x}) * K_{0.5,x}$$

$$SAR_{x} - \text{the specific activity ratio in animal x;}$$

$$SAR_{x} - \text{the specific activity of bound hydrogen (BH) in the predator}$$

$$(kg BH kg^{-1} fw)$$

$$M = SAR_{x} * K_{0.5,x} * \frac{SA_{pred}}{1111}$$

$$SAR_{x} - \text{the specific activity of bound hydrogen (BH) in the predator}$$

$$(kg BH kg^{-1} fw)$$

$$M = -mass of free hydrogen (kg) in 1 m^{3} of water$$

 $SA_{pred} = 0.06*Dryf_{pred}$ (excepting the fish fat and depending on dm fraction of the predator)

For the fish fat, we recommend $SA_{pred} = 0.08*Dryf_{pred}$

OBT dynamics in zooplankton

K_{0.5} depends on its growth rate and temperature



- $K_{0.5_0}$ OBT biological loss rate at the reference temperature (d-1); V $\,$ the zooplankton volume (μm^3)
- for different species of zooplankton \rightarrow V = 10 \div 104 μm^3 ;
- $K_{0.5 0}$ varies between 0.19 and 0.7 day⁻¹, with an average of 0.3 day⁻¹;
- For the present case, we choose the minimum value, appropriate for large zooplankton:

 $K_{0.5} = 0.19 * 1.06^{(T-20)} \rightarrow$ We introduced the temperature dependence

dm fraction varies between 0.07 and 0.2 and we used 0.12 as a default value.

OBT dynamics in zoo benthos

- The benthic fish consume macrointervertebrates and especially, aquatic insects' larvae of Diptera order.
- The most widespread ones are those from *Chironoma* family, which has 2 6 life cycles per year.
- Chironoma larvae:

growth rate - 0.05 day⁻¹ and a respiration rate - 0.01 day⁻¹ (Heling 1995);

 $K_{0.5} = 0.06 \text{ day}^{-1}$ (Heling 1995); $K_{0.5} = 0.2 \text{ day}^{-1}$ (CASTEAUR); $K_{0.5} = 0.1 \text{ day}^{-1}$ (the present application - average value)

All the previous values for $K_{0.5}$ correspond to an average water temperature of 12 °C.

• The small molluscs and crustaceans have a very large variability and the calculations of their $K_{0.5}$ must be adapted to different cases.

• Molluscs:

 $K_{0.5} = 0.02 \text{ day}^{-1}$ for a mass of 1 g (Heling and Galeriu, 2002) $K_{0.5} = 0.005 \text{ day}^{-1}$ for 30 g of soft tissue (Heling and Galeriu, 2002) $K_{0.5} = 0.017 \text{ day}^{-1}$ (Heling 1995)

• Crustaceans:

 $K_{0.5} = 0.007 \text{ day}^{-1}$ (Heling and Galeriu, 2002)

 Based on experimental data for the growth rate and the energy content of *Mytilus* edulis soft tissue (2386 J per g wet tissue), it is given the following relationship (Sukhotin et al., 2002):

$$K_{0.5_0} = 0.024 * W^{-0.246}$$

- **Elliptio complanata** total mass 90 g (40 g wet mass) $K_{0.5} = 0.02 \text{ day}^{-1}$ (Mussel uptake scenario, 2008)
 - For the food chain modelling, we are interested in molluscs and crustaceans consumed by humans and in various species of zoo benthos consumed by fish.
 - The model has two separate compartments.
 - For human consumption we included mussels and crabs with large mass and the model parameters are adapted to approximate this mixture.
 - By default, we use a value $K_{0.5} = 0.007 \text{ day}^{-1}$, but the users must adapt this value to their specific cases.
 - As fish prey, we considered the zoo benthos with smaller mass and $K_{0.5} = 0.04 \div 0.1 \text{ day}^{-1}$
 - There is also a temperature dependence which is introduced as a multiplicative factor:

$$K_{0.5} = F(T) * K_{0.5_0}$$

$$F_{molluscs,crustaceans}(T) = \frac{0.043 - 0.0159 * T + 0.003 * T^{2} - 5.3 * 10^{-5} * T^{3}}{0.51}$$

F(T) – the temperature correction function;

 $K_{0.5 0}$ – the OBT biological loss rate at standard temperature (day⁻¹)

OBT dynamics in fish – Fish bioenergetics models (BEMs)

- Too few experimental data for K_{0.5} of OBT
- In absence of the experimental data, we used models based on bioenergetics.
- Experimentally demonstrated that the mass dependence of fish basal metabolic rate is a combination between the tissue specific respiration rate and tissue relative size (Oikawa and Itazawa, 2003).
- We used the same approach as for mammals (*i.e* the energy metabolism approach) (Galeriu et al., 2009).
- 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 the following:

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

- W fish mass (g fw);
- t time (day);
- C consumption (g prey g^{-1} fish day⁻¹);
- R respiration or losses through metabolism (g prey g⁻¹ fish day⁻¹);
- S specific dynamic action or losses because of energy costs of digesting food (g prey g⁻¹ fish day⁻¹);
- F egestion or losses through faeces (g prey g⁻¹ fish day⁻¹);
- E excretion or losses of nitrogenous wastes (g prey g^{-1} fish day⁻¹);
- P egg production or losses through reproduction (g prey g⁻¹ fish day⁻¹);
- cal_{p} , cal_{f} caloric equivalents of pray (J g^{-1}) and fish (J g^{-1}), respectively

Equation for consumption:

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

- C 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); $C_{max} = aW^b$ with a, b allometric coefficients for fish;
- p the proportion of maximum consumption;
- fc(T) a temperature dependent function
- Respiration measured as oxygen consumption converted to consumed prey, by knowing the energy equivalent of oxygen (13560 J $g^{-1} O_2$) and the prey energy density.
 - depends on temperature, fish mass (allometric function) and activity:

$$R = a_r W^{br} f_r(T) A CT * conv$$

- R respiration (g prey g⁻¹ fish day⁻¹);
- a_r , b_r allometric coefficients (a_r is usually given in units of O_2 consumption per g fish and unit time);
- $f_r(T)$ temperature function of respiration;
- ACT activity multiplier depending on fish average swimming speed;
- conv oxygen consumption converted to consumed prey (13560 J g⁻¹ O₂ calp⁻¹)
- In many applications, the specific dynamic action (S), the egestion (F), and the excretion (E) depend on consumption, as an overall fraction (ε), and RGR is given by:

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

• The OBT biological loss rate K_{0.5} :

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

We must note the effect of growth dilution (RGR) and maintenance (respiration) rate.

Dissolved organic tritium (DOT)

• The model described is based on the assumption that the OBT SA in fish is directly linked with the HTO in water or the OBT in fish food \rightarrow fully valid if the water contamination is due only to an initially HTO source $\rightarrow CF \leq 1$

CF=concentration per unit mass of biota at equilibrium / dissolved concentration per unit volume in ambient water

- CFs in marine biota at Cardiff Bay (UK) much higher (McCubbin et al., 2001; Williams et al., 2001)
 - flounder
 - mussels
- CFs ≥4 × 10³ (fw equivalent) → 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 authorised releases of wastes to the Bristol Channel from the Nycomed-Amersham (now GE Healthcare) radiopharmaceutical plant at Whitchurch, Cardiff, UK
- The extremely high CFs can't be explained by analytical errors (Hunt et al., 2010)
- Advanced hypotheses:
 - concentration of organic tritium by bacteria and subsequent transfer in the food chain;
 - ingestion of contaminated sediment;
 - ingestion of contaminated prey;
 - direct uptake of DOT from the sea water;
 - bioaccumulation occur via a pathway for the conversion of the organic compounds labelled with dissolved ³H into particulate matter (via bacterial uptake / physico-chemical

sorption) and the subsequent transfer to the foodchain (McCubbin et al., 2001) \rightarrow not valid, because monitoring data on sediment and suspended matter compared with data on tritium in benthic fauna show that the ingestion of sediment or particulate matter is not a reasonable explanation

- Phytoplankton can selectively assimilate dissolved organic carbon (DOC) from water (Neilson and Lewin, 1974) and the laboratory experiments show the same fact for tritiated organics (Strack et al., 1983).
- The same fact is true for mussels (Jorgensen 1983).
- In a review about amino acids transport in marine invertebrate (Wright and Stephens 1982), it is pointed out that all the soft-bodied marine invertebrates have the ability to accumulate some radioactive labelled amino acids and most of them have a net influx.
- The transport process is transepidermal in nature and can be described by the Michaelis-Menten kinetics.
- Transport rates are related to levels of metabolic energy and the accumulated substances are available to general metabolic pathways.
- It is important to note that this transport process is rarely observed in freshwater invertebrates.
- The Michaelis-Menten kinetics is described by the following equation:

Ji

$$J^{i} = \frac{J^{i}_{\max}[S]}{K_{t} + [S]}$$

- the influx rate of substrate (μ mol g⁻¹hour⁻¹);
- J_{max}^{i} the maximal rate of influx (µmol g⁻¹hour⁻¹);
 - [S] the concentration of substrate ($\mu \mod L^{-1}$);
 - the concentration of substrate at which the influx is K, one-half of maximal value ($\mu \mod L^{-1}$)

When the concentration of substrate is much smaller than the half saturation constant, the previous $V = \frac{J_{\max}^{i}}{K_{t}}$ equation is simplified and it can define an uptake rate:

Experimental validation:

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- marine unicellular algae (*Dunaliella bioculata* and *Acetabularia mediteranea*) grown in 10 different tritiated organic solutions of amino acids (Strack et al., 1983). The organics have been supplied to the algae culture for 30 min. and the concentration ratio (fw) varied between 0.17 and 122 (3 orders of magnitude). The maximum concentration ratio was for adenine in *Dunaliella bioculata*.
- In a separate experiment (Strack et al., 1980), the Michaelis –Menten kinetics was analysed for adenine uptake in *Dunaliella bioculata* and the uptake rate, V, was estimated at 4800 L g⁻¹ dm hour⁻¹.
- We introduced the direct uptake of DOT in the dynamic eqs. for phytoplankton and consumers:

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

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

 C_{W} - HTO concentration in water (Bq m⁻³); C_{DOT} - the dissolved organic tritium concentration (Bq L⁻¹); V_{DOT} - the uptake rate of DOT (L kg ⁻¹fw day⁻¹)

RESULTS

OBT in phytoplankton and zooplankton

- The OBT dynamic equation for phytoplankton was successfully tested with many experimental data (Heling and Galeriu, 2002).
- There are no direct data for OBT biological loss rate in zooplankton, but the dynamics of OBT concentration in a specie of zooplankton (*Artemia salina*) (Komatsu et al., 1981) is consistent with the generic value used in the present model

OBT in molluscs and crustacean

- Due to the large variability of respiration rate in invertebrates (Brey 2010) and the paucity of the experimental data for OBT biological loss rate (Heling and Galeriu, 2002), it is not possible to use a single robust, generic value.
- The experimental data for *Mytilus edulis* are closely reproduced by the present model, but only those for marine clams (*Mya arenaria*) are predicted within a factor 2-3, using the available data for respiration.

Results - OBT dynamics in fish

- We selected only the cases with experimental verification
- Pacific herring (Megrey et al., 2007):

- the model parameters have been obtained mainly from the laboratory experiments, and there have been considered the realistic seasonal changes in water temperature and prey availability.

- Prey availability (consumption fraction) and temperature dynamics are the major factors influencing the dynamic of OBT biological loss rate



Temperature and prey availability dynamics (consumption fraction is multiplied by 50, in order to have a similar scale with that one for temperature) for Pacific herring For fish masses (in consumption mass range) the OBT biological half time varies between 60 and 170 days



Mass and OBT biological loss rate dynamics (multiplied by 10⁴, in order to have a similar scale with that one for mass) for Pacific herring

For another pelagic fish, the Pacific saury (*Cololabis saira*) near the Japanese coast (Ito et al., 2004 the range of OBT biological half times for a similar mass is 40 - 60 days, due to differences in metabolism, water temperature and prey availability.

<u>Roach</u>

• Laboratory experiments for consumption and respiration have been done for a mass range of 1.2 - 300 g and a temperature range of 5 – 23 ° C, providing improved parameters (Hoelker and Haertel, 2004),

• The improved parameters (HH), as well as an old set of parameters (Horppila and Peltonen, 1997), have been used to assess the OBT biological half time.

Temperature (°C)	OBT biological loss rate (day-1)				
	Maintenance ration ^a		Half maximum feeding rate ^a		
	НР ^ь	HH¢	НР ^ь	HH¢	
5	0.0011	0.0016	0.0035	0.003	
10	0.002	0.0031	0.0065	0.0057	
15	0.0036	0.0026	0.0114	0.011	
20	0.0063	0.0061	0.019	0.021	
25	0.01	0.008	0.028	0.042	

OBT biological loss rate of roach for two feeding regimes

^a Feeding regimes

^b Old set of parameters (Horppila and Peltonen, 1997)

^c Improved set of parameters (Hoelker and Haertel, 2004)

The experimental data for larger fish are not reported in literature, but there is work in progress (Kim SB Personal communication 2010) for the rainbow trout with a mass of about 400 g and the water temperature of 15 °C.

For this case the present model predict an OBT biological loss rate of about 40 days⁻¹.

The present model results suggest that in field conditions, the major factors influencing the OBT biological loss rate are temperature and prey availability.

EMRAS Mussel Scenario

- First phase the mussels have been relocated form a low background area to a highly tritium contaminated lake and the uptake of tritium, including OBT, had been studied for 84 days (uptake phase).
- Second phase mussels grown in a contaminated lake have been relocated in clean water and tritium depuration had been studied for 117 days (depuration phase).
- The AQUATRIT model, using its earlier set of parameters (Galeriu et al., 2005) (*i.e* $K_{05,0}$ =0.022 day⁻¹).
- For the uptake phase, the model under predicted the experimental data by a factor up to 5 for the first 6 days and for the later phase of the experiment, the model's predictions were close with the experimental data.
- For the depuration phase, the model largely under predicts the experimental data, by a factor up to 15.
- Decreasing the biological loss rate used in the model at 0.004 day⁻¹, the agreement between the model's predictions and the data is good, but this contradicts the uptake parameters' values.
- It seems that a simple model with only one compartment is not appropriate for such scenario with abrupt changes of the environmental conditions.
- A two compartment model was used by one of the participants with good performances for depuration phase, but with poorer performances for the uptake phase.
- The best solution seems to consider the stomach content and a single OBT compartment.
- When the mussels are moved to the contaminated area, the stomach is saturated with contaminated food in few hours and the food gradually passes to rest of the body. Considering a stomach representing a fraction of 0.3 from the whole body and an OBT biological loss rate of 0.011 day⁻¹, both the uptake and the depuration phases are successfully modelled (the model's prediction is up to a factor 2).

Danube ecosystem

- A scenario for a hypothetical accidental tritium release in Danube of 3.7 PBq for 6 hours is analysed, using the information about the Danube River and Delta ecosystem gathered in a recent environmental impact assessment
- Seasonal changes in water flows and temperature were considered and the model parameters were adjusted for predominant species of phytoplankton, zoo benthos, and fish.
- For the modelling purposes, we considered a carp with a mass of about 1000 g and with the bioenergetic parameters as those given in AQUATOX (Park and Clough, 2009), but adjusting the respiration in order to reproduce the recent data (Ohlberger et al., 2005).
- Carp diet contains 40 % benthic algae, 10 % zooplankton, and 50 % zoo benthos.
- As a representative predator fish we selected pike and zander. For both fish we selected the recent parameters.
- For this application roach was selected as prey fish, with the mass of about 100 g and robust model parameters have been selected (Hoelker and Haertel, 2004). Roach diet is 60 % zooplankton, 30 % zoo benthos and 10 % benthic algae.

Fish growth for local conditions and for two years



Fish OBT biological loss rate



The dynamics of OBT concentration in different aquatic organisms, considering a tritium release on August 1 and a river flow of 6000 m³s⁻¹,



• In practice, an incident with tritium loss in Danube River can occur any time and it will be useful to understand the seasonal effect of the release impact on human ingestion coming from fish. Across the years, the Danube River's flow and temperature vary and for the same release of 3.7 PBq of tritium for 6 hours, the fish contamination varies also

Date of release	River flow (m ³ s ⁻¹)	River temperature (°C)	Ingested activity of fish (Bq) ^a	% OBT ^b
February 15	3000	3	22844	3
April 15	5000	10.5	14348	7.3
May 15	3500	17	21831	13
July 15	1500	24	63377	30
September 15	1000	20	92430	28
October 15	1500	15	53790	17.5
December 15	1500	5.5	46415	4.4

^a 0.5 kg of a mixture of carp and zander

^b The percentage of OBT coming from the ingested fish activity

Water temperature has a large influence on the OBT content in fish and the highest impact is in late summer (September 15).

DOT

For the Cardiff case it should be noted that the tritated waste from GE Healthcare (former Amersham) includes not only the HTO and the by-product, but also the high bio available tritiated organic molecules (*i.e.* hydrocarbons, amino acids, proteins, nucleotides, fatty acids, lipids, and purine / pyrimidines).

For the model application, the input data as: the annual average of total tritium and organic tritium releases from GE Healthcare, tritium concentration in sea water and the monitoring data for mussel and flounder have been taken from literature



Figure 1: Record of discharges of tritiated waste from Amersham plc into the Severn Estuary

Using the available input data, the model successfully predicts the trend for tritium concentration in mussels and flounders

Comparison between model results and monitoring data for mussels (*Mitilus Edulis*)



Comparison between model results and monitoring data for flounder (*Platichthys flesus*)



• Intensive monitoring activity was carried out in 1999-2000 (Leonard et al., 2001) and shows that the pelagic fish have a 10 fold lower tritium concentration than the benthic flounder.

- This is also seen in the model outputs (it used herring as pelagic fish).
- The above results, despite parameters uncertainty, demonstrate that the direct uptake of DOT in phytoplankton and invertebrates is the most probable explanation for the OBT dynamics in fish species in Cardiff area.

The previous model results must be considered with some restrictions:

- seasonal variability of release (including organic tritium) is not known and organic species contribution can vary across the year the detailed information is not available;
- DOT transfer to marine phytoplankton and invertebrates is less documented in relation with species and chemical forms of organic tritium uncertainty in model parameters is high and difficult to assess;
- monitoring results are relatively few with a spread of the values attaining a factor of at least 3 among the providers and this affects the relevance of the annual average.

CONCLUSIONS

- In the late 1980, the aquatic pathways after releases of tritium (HTO) were not considered of relevance (Blaylock et al., 1986) and simple models were used based on specific activity approach.
- The occurrences of high concentration factors in Cardiff area generate debate and public concern for the development of nuclear pharmaceutical production.
- The present model intends to be more specific than a screening model, including a metabolic approach and the direct uptake of DOT in marine phytoplankton and invertebrates.
- The high concentration factors found in Cardiff area are not a general problem of nuclear industry. The Cardiff case reflects a specific biological process in marine invertebrates and the consequences were ignored in the past.
- In order to have a better control of tritium transfer into the environment, not only tritiated water must be monitored, but also the other chemical forms and mainly, OBT in food chain.

