EMRAS Tritium/C14 Working Group

THE PERCH LAKE SCENARIO

Final Report
May 2006

1. SCENARIO DESCRIPTION

This scenario is based on data collected in Perch Lake, a small, shallow freshwater lake located within the borders of AECL's Chalk River Laboratories in northeastern Ontario (Figure 1). The lake contains elevated levels of tritium due to long-term discharge from nearby waste management areas. The tritium forms a well-defined subsurface plume that discharges into the lake through sediments and a stream (Inlet 2 in Figure 1). Inlet 1 shows slightly elevated levels of tritium but Inlets 3, 4 and 5 are all uncontaminated.

Tritium concentrations were measured in samples of air, lake water, sediments, aquatic plants (algae, bladderworts, hornworts and cattails) and animals (clams, bullheads and pike) collected in the summer and fall of 2003. Bladderwort and hornwort (hereafter referred to collectively as worts) are both unrooted plants that are completely submerged and obtain their nutrients from the water. These two species were composited for analysis. The cattails are rooted in the top 5-10 cm of the sediments, from which they draw their nutrients. They extend above the water into the air, and the submerged and emergent parts were analysed separately. Bullheads are omnivorous, benthic fish and pike are larger piscivores. Both types of fish likely move throughout the lake, eating other fish and invertebrates. The fish samples were divided into three parts (flesh, head and internal organs), each of which was analyzed separately.

The air, water, sediment and plant samples were taken primarily from three locations: at S1, located near Inlet 1; at S2 near Inlet 2; and at S3 near Inlet 3 (Figure 1). A few samples were also taken at S4 near Inlet 4 and near the outlet of the lake. Some water samples were collected from the surface of the lake and others at depth near the bottom. Most of the plant and sediment samples were collected from shore at the edge of the lake. Some of the water samples were also taken close to shore but others were collected by boat 50-100 m offshore, as were algae. Fish tend to feed on the east side of the lake and were caught in two extended areas on either side of the outlet, whereas clams were harvested between Inlet 3 and the outlet. Most samples were collected three times during the summer and fall of 2003 (May 29-30, July 28-29 and October 1-2). Additional measurements of water concentrations were made in early November. Air concentrations were measured only in August and October as monthly averages and algae and clams were not available in October. Replicate samples were taken in some cases. All samples were analyzed for their HTO content, and OBT concentrations were determined for the sediments, plants and animals.

Given the measured HTO concentrations in water, sediments and air, participants in the scenario were asked to calculate

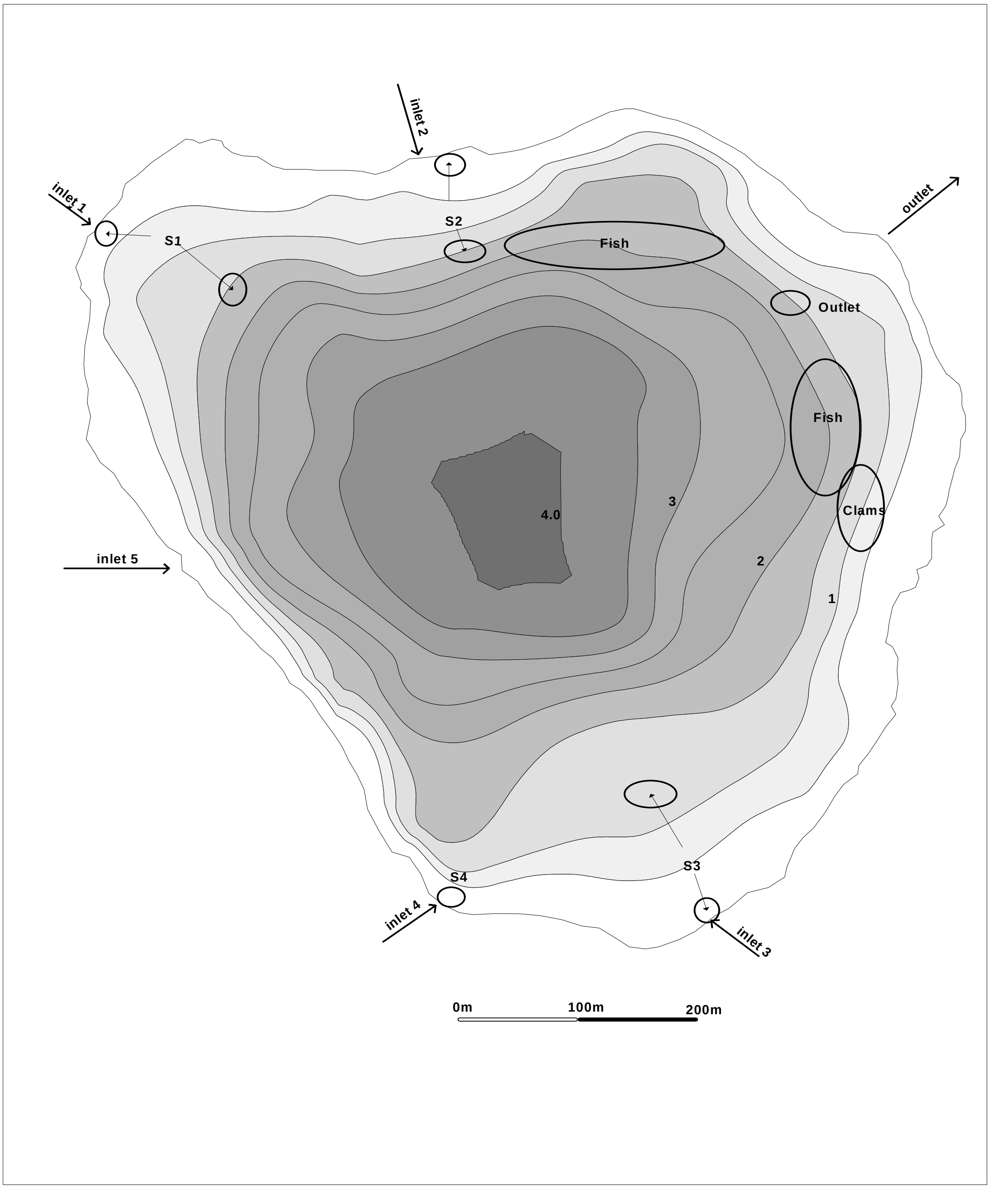

Figure 1. Map of Perch Lake showing inlets, the outlet, depth contours in m and the sampling locations.

(i) HTO and non-exchangeable OBT concentrations in cattails and worts for the May sampling period for the near-shore portions of sites S1, S2 and S3. For cattails, concentrations were requested for both the above water and below water parts of the plant.

(ii) HTO and non-exchangeable OBT concentrations in algae for the May sampling period for the offshore portions of sites S1, S2 and S3.

(iii) HTO and non-exchangeable OBT concentrations in clams, bullheads and pike for each of the three sampling periods. For bullheads and pike, concentrations were requested in head, flesh and internal organs (liver, gonads, stomach and intestines).

(iv) non-exchangeable OBT concentrations in sediments for the May sampling time for the near-shore portions of sites S1, S2 and S3.

(v) 95% confidence intervals on all predictions in (i) - (iv).

The data included in the scenario represented a relatively small subset of all the data collected in the experimental program. The full data set has been presented and analyzed by Kim et al. (2004). The full scenario description is given in Appendix A.

2. OBSERVATIONS

2.1 Measured Concentrations: Measured HTO concentrations in air moisture, lake water and sediment water are shown in Table 1. These are the concentrations that were supplied to the participants to drive their models. Observed HTO and OBT concentrations in plants, animals and sediments, which were the endpoints of the scenario, are given in Tables 2 and 3. The OBT concentrations are given in units of Bq L^{-1} of combustion water.

Counting errors in the HTO concentrations for lake water, plants and aquatic animals were generally less than 2%, but reached about 10% in some cases. These errors likely represent the full uncertainty for the lake water samples, which are easy to collect and analyse. Additional differences of perhaps 30% would be expected from sample to sample in plants and animals due to natural variability. Counting errors in the sediment concentrations were larger, reaching up to 25% in some cases, and the total uncertainty may be somewhat greater because of difficulties in keeping the sediment pore water distinct from the lake water. Uncertainties in air concentrations arose due to counting errors and the performance of the samplers, and are estimated to be about 30%. Counting errors for OBT concentrations were usually less than 5% but additional uncertainty arose due to difficulties in removing exchangeable OBT from the samples and in the combustion process. The total uncertainty in the OBT measurements is estimated to be about 20%, although greater variation must be expected among individual plants and animals.

Table 1. Measured HTO concentrations in water, sediment water and air moisture

Month	Compartment	HTO Concentrations ($Bq\ L^{-1}$)				
		S1	S2	S3	S4	Outlet
May	Surface water - offshore	4350	5450	4730		
	Sediment water - offshore	4730 3330 3830	10890 13570 13210	1320		
July	Surface water - offshore - from shore near inlet	4640 4150	4590 3330	4620 3800	 91	4660
	Bottom water - offshore* - from shore near inlet‡	4480 3900	4460 2570	4420 3580		4620
	Sediment water - from shore near inlet	2300	7120	70		
Oct	Surface water - from shore near inlet	2030	9290	139		
	Bottom water - from shore near inlet‡	2080	9190	113		
	Sediment water - from shore near inlet	1500 1650	7420 4550	84		
	Air - August - October	740 660	1970 1770	510 260		
Nov	Surface water - offshore	3840	5270	3770		
	Bottom water - offshore*	3480	9350	3770		

* collected at a depth of about 1.5 m; ‡ collected at a depth of about 0.4 m

Table 2. Observed HTO and OBT concentrations in plants and sediments in May

Compartment	HTO (Bq/L)			OBT (Bq/L combustion water)		
	S1	S2	S3	S1	S2	S3
Cattails - emergent	1970	8080	1180	1500	4100	971
- submerged	3390	9760	1360	2120	3760	655
Worts	4680	6020	4520	2500	3230	1580
Algae	6630	5490	4990	2610	3200	2410
Sediments	See Table 1			1960	2970	488

Table 3. Observed HTO and OBT concentrations in clams and fish

Fish type	HTO (Bq/L)			OBT (Bq/L combustion water)		
	May	Jul	Oct	May	Jul	Oct
Clams	5750	4100	-	3270	3810	-
Bullheads - head	5270	4070	3230	3820	3160	4110
- flesh	5310	4050	3230	3970	3480	3820
- internal organs	5240	4040	3620	3610	3340	3520
Pike - head	5120	4100	3470	3630	4050	4480
- flesh	5020	4130	3460	3950	3710	4500
- internal organs	5170	4100	3510	3780	3460	4610

2.2 Analysis of Observations: The following conclusions can be drawn from an analysis of the full Perch Lake data set (Kim et al. 2004).

HTO Concentrations

- Within experimental uncertainty, the HTO concentrations in the aqueous parts of algae and worts are indistinguishable from the HTO concentrations in the water surrounding them (plant/water = 0.94 ± 0.27; n = 12; 1 outlier ignored).

- HTO concentrations in the submerged parts of cattails are the same as HTO concentrations in sediment pore water (plant/sediment = 1.06 ± 0.29; n = 9).

- HTO concentrations in the emergent parts of cattails (C_{ec}) are well predicted by the equation $C_{ec} = 1.1 (C_{am} + C_{sed})/2$, where C_{am} and C_{sed} are the HTO concentrations in air moisture and sediment water (predicted/observed = 1.03 ± 0.21; n = 9). The factor 1.1 is the ratio of vapour pressures between water and HTO, and is introduced by analogy with the model for terrestrial plants (Murphy 1984).

- HTO concentrations in clams are the same as HTO concentrations in bottom waters averaged over all offshore locations (clam/water = 1.05 ± 0.14; n = 2).

- HTO concentrations in bullheads are indistinguishable from the HTO concentrations in bottom waters averaged over inshore and offshore locations (fish/water = 0.96 ± 0.06; n = 3).

- HTO concentrations in pike are the same as HTO concentrations in water averaged over the entire lake, including surface and bottom water and onshore and offshore water (fish/water = 0.96 ± 0.04; n = 3).

- HTO concentrations in the flesh, head and internal organs of the fish show no significant differences.

OBT Concentrations

- OBT concentrations in the combustion water of algae and worts are proportional to the HTO concentrations in the aqueous part of the respective plants, averaged up to the time of sampling. The mean observed OBT/HTO ratio is 0.48, with a standard deviation ± 0.08 (n = 13).

- OBT concentrations in all parts of cattails are proportional to the HTO concentrations in the emergent part of the plant, averaged up to the time of sampling. The mean observed OBT/HTO ratio is 0.70, with a standard deviation ± 0.19 (n = 18).

- OBT concentrations in clams, bullheads and pike are proportional to the HTO concentrations in water, averaged spatially over the locations accessed by the species in question and temporally up to the time of sampling. The mean observed OBT/HTO ratio is 0.79, with a standard deviation ± 0.09 (n = 8).

- OBT concentrations in the flesh, head and internal organs of the fish show no significant differences.

- OBT concentrations in sediments are about 60% of the OBT concentration in plants (n = 9).

The OBT concentrations in all plants are less than the corresponding HTO concentrations primarily because of isotopic discrimination in the formation of OBT. Similarly, animal OBT concentrations are less than HTO concentrations because of metabolic processes that tend to convert OBT to HTO.

3. MODEL DESCRIPTIONS

Eight participants submitted results for this scenario (Table 4). All participants treated the scenario as a blind test of their models and submitted results before the observed concentrations were made known to them.

The Perch Lake scenario tested models that predict tritium concentrations in an aquatic ecosystem subject to a continuous release of HTO. It was a fairly simple scenario in the sense that releases to the lake have been going on for many years at roughly the same rate, and tritium concentrations in various parts of the ecosystem are likely to be in equilibrium. However, the scenario showed a number of complicating factors. Since tritium enters the lake through the sediments, concentrations in sediment pore water are generally higher than those in lake water, which in turn are higher than those in air. The sediments themselves show a spatial gradient in concentration, with larger values in the parts of the lake closest to the subsurface tritium plume.

Table 4. Participants in the Perch Lake Scenario.

Participant	Affiliation	Designation in text
A. Golubev	VNIIEF, Russia	VNIIEF
F. Siclet	EDF, France	EDF
M. Saito	Safety Reassurance Academy, Japan	SRA
F. Baumgärtner	Technische Universität München, Germany	BioM
D. Galeriu	IFIN-HH, Romania	IFIN
Japanet*	Japan	J
P. Marks	GE Healthcare, U.K.	GE
T. Nedveckaite	Institute of Physics, Lithuania (LIETDOS_W model)	L

***** Participants from NIRS, Ibaraki University, Kumamoto University, Toyama University and Kyoto University, led by K. Miyamoto and Y. Inoue from NIRS

Water concentrations in a narrow zone close to shore may be higher or lower than those in the main body of the lake, depending on the tritium concentration in the water in the streams flowing into the lake. Concentrations in sediments, lake water and air all varied gradually with time during the study period. Finally, the sediments were composed of a mixture of sand and gyttja (decomposing organic matter), with proportions varying through the lake. In the face of this variability, the modelers had to make a number of decisions: which source compartments (sediments, water or air) contributed tritium to the plants and animals for which predictions were requested; how to average over space to reflect the concentrations seen by the fish, which move freely throughout the lake; how to average over time when calculating concentrations of OBT, which has a long biological half life in all organisms; and how to estimate the required water concentrations when the relevant data was missing or incomplete in the scenario description.

Once these decisions were made, most modelers assumed the HTO concentration in a given endpoint was equal to the average water concentration in the source compartment(s). The exception was L, which assumed that HTO concentrations in plants and animals were slightly lower than in water. The OBT concentration in a given endpoint was generally based on the corresponding HTO concentration, with some allowance made for isotopic discrimination in the case of plants and metabolic processes in the case of animals. The IFIN model generated predictions for each plant type that were representative of a whole lake average, using data from Cornett (1989) to augment the information in the scenario description on bottom water concentrations. Two participants (IFIN and GE) used dynamic models to estimate OBT concentrations in algae and all animals, and a third model (EDF) took a similar approach for fish. In each case, these models took account of the growth rate of the animal, ingestion and excretion rates and internal metabolic/catabolic processes to describe the incorporation of OBT in the animal and the conversion between OBT and HTO. The participants showed considerable variability in their approach to modeling sediments.

The BioM model gives different OBT endpoints than those of the other models, predicting the concentration of buried tritium rather than the tritium traditionally considered to be organically (or carbon) bound. Buried tritium is tritium in exchangeable positions that is not removed by the conventional rinsing process. It consists primarily of tritium in large molecules that becomes hidden from the effects of washing when the free water in the sample is extracted by freeze drying or azeotropic distillation. A smaller part consists of tritium in hydrate bonds that is similarly not removed by washing, but this is not accounted for in the model. Buried tritium appears as part of the experimental yield when the sample undergoes traditional analysis for OBT, but is converted to HTO as soon as it is ingested. BioM calculates the concentration of buried tritium from the HTO concentration in the sample assuming a two-step exchange process and taking into account the proportion of carbohydrates, proteins and DNA in the tissues. The difference between the observed OBT concentration and the predicted buried tritium concentration gives the organically bound (or carbon bound) tritium concentration for the BioM model, if the tritium in the hydration shells is neglected.

The participants estimated the uncertainties in their predictions using very different methods. One modeler (L) carried out a rigorous Monte Carlo uncertainty analysis using lognormal distributions for the HTO concentrations in water and the bioaccumulation factors. At the opposite end of the spectrum, IFIN used expert judgment, arguing that the main source of uncertainty was the lack of detailed information on HTO concentrations in water as a function of time and space in the lake. J also used expert judgment, setting the uncertainty in a given endpoint at ±20% of the water concentration used to predict that endpoint. Between these extremes, EDF, SRA and BioM used the variability in the observed water concentrations as the basis for their uncertainty estimates but even here the individual approaches were quite different. EDF carried out a perturbation analysis to estimate uncertainties in most OBT concentrations and used the range of predictions from different conceptual models to arrive at the uncertainty in sediment OBT concentrations.

Details of the models are introduced in the following sections as they are needed to explain the results. Full model descriptions are given in Appendix B.

4. COMPARISON OF PREDICTIONS AND OBSERVATIONS

4.1 **Overall Results:** When the predictions of all the models were averaged for a given endpoint, the results agreed well with the corresponding observation (Table 5). The mean predictions lay within 30% of the observations for all HTO concentrations. Similar agreement was found for the OBT concentrations, except in the case of sediments and the underwater parts of cattails, where concentrations were overestimated by factors of 2.3 and 1.7, respectively. Most participants derived the OBT concentrations in submerged cattails from the HTO concentration in the same part of the plant. This leads to overestimates since the data suggest that OBT is formed in the emergent parts and translocated to the submerged parts. HTO concentrations in the emergent parts are low because of losses to the relatively uncontaminated air, and the OBT produced will be correspondingly low.

OBT in sediments is expected to arise from decaying plant and animal material deposited on the lake bottom, with the greatest contribution coming from plants. The sediment concentration was observed to be lower than the concentration in plants by about a factor of 2. This could be due to the increasing age of the organic material in deeper parts of the sediments, which could result in decreasing activity due to decay or breakdown of OBT as the organic matter decomposes. Most participants assumed concentrations were equal in plants and sediments and overestimated the sediment concentrations.

Table 5. Comparison of predictions averaged over all models to the observations for each endpoint

Endpoint	Ratio of mean prediction to observation	
	HTO	OBT
Algae	0.92	1.2
Worts	0.83	1.3
Submerged cattails	1.2	1.7
Emergent cattails	1.3	1.4
Bullhead flesh	1.1	0.87
Pike flesh	1.0	0.71
Clams	0.95	0.91
Sediments	*	2.3

* HTO concentrations in sediments were given as part of the scenario description

The results shown in Table 5 indicate that, with the exception of OBT in sediments and the underwater parts of cattails, the modelers as a group have a good conceptual understanding of the behaviour of tritium in the Perch Lake ecosystem and can predict HTO and OBT concentrations that, in an average sense, agree well with the observations. However, the scatter in the predictions of individual models was substantial. Table 6 shows the ratio of the largest prediction to the smallest for each endpoint in the scenario. The ratios range from 1.7 to more than 100 and are larger for OBT than for HTO. The largest values occur for sediments and the emergent parts of cattails, where the modelers showed the greatest divergence in their conceptual approaches.

Table 6. Range of model predictions for each endpoint

Endpoint	Ratio of highest to lowest prediction	
	HTO	OBT
Algae	1.8	2.6
Worts	1.7	4.1
Submerged cattails	93	103
Emergent cattails	6.5	103
Bullhead flesh	3.3	6.1
Pike flesh	2.6	7.8
Clams	2.8	5.2
Sediments	*	97

*HTO concentrations in sediments were given as part of the scenario description

Results for each scenario endpoint are discussed in turn below. For the plant and sediment endpoints, emphasis will be given to sampling site S3, where spatial gradients are expected to be smallest. For the animal endpoints, the discussion will focus on the results for July, for which there is the largest amount of information on HTO concentrations in water and sediments.

4.2 Algae: Predictions for the HTO concentration in algae at sampling site S3 in May are compared with the observation in Figure 2. Because algae is completely submerged in water, and because tritium in so mobile in the aqueous phase, the HTO concentration in algae collected at a given time and place is expected to equal the local water concentration. This expectation was borne out in the analysis of the full Perch Lake dataset (Kim et al. 2004) and is evident at site S3 in May, where the HTO concentration in algae (4990 Bq/L) was within 5% of the water concentration (4730 Bq/L). Five of the participants (EDF, SRA, BioM, IFIN and J) assumed the HTO concentration in algae was equal to the local water concentration and so achieved a good result (Figure 2). GE is a dynamic model and overestimated the observation slightly, but even here the prediction lay within the uncertainty bounds of the data. VNIIEF assumed that the algae were in equilibrium with water concentrations averaged over surface and bottom layers and underestimated the observation It is not clear if this underprediction is significant because the modeler did not estimate the uncertainties in his concentrations. Participant L did not submit predictions for algae.

The results for Site S2 followed the same pattern as for S3, with all models but VNIIEF producing results in good agreement with the observation. At S1, all of the models predicted a concentration in algae close to the water concentration (4350 Bq/L) but for some reason the observed concentration in algae at this site was substantially higher at 6630 Bq/L.

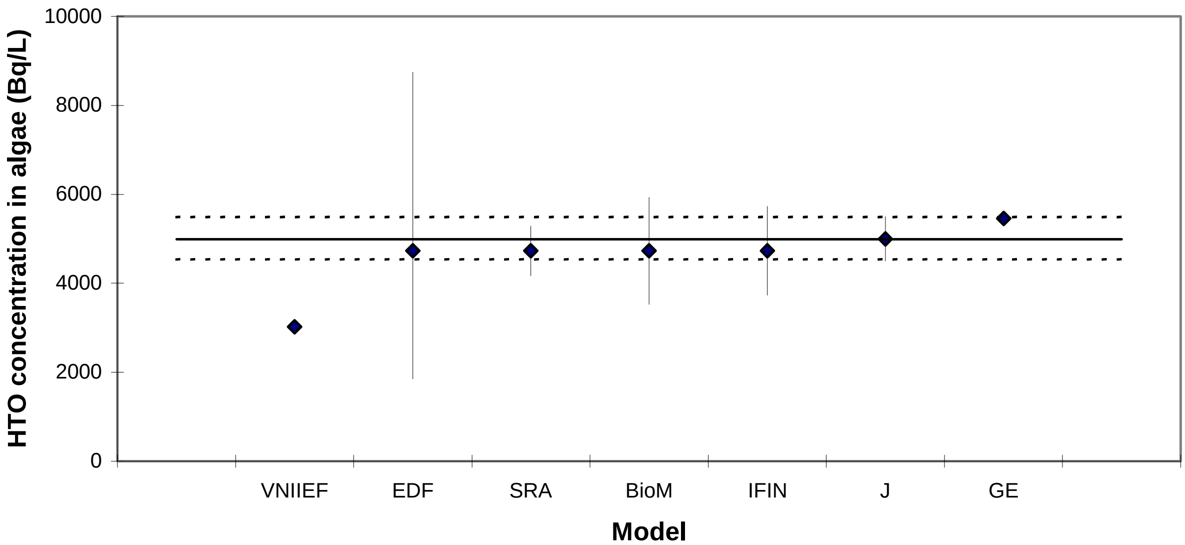

Figure 2. HTO concentrations in algae at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates and L did not submit results for this endpoint. The offshore water concentration at S3 in May was 4730 Bq/L.

Because of the slow turnover rate of OBT in algae and other plants, OBT concentrations are expected to depend on the plant HTO concentration averaged over the few weeks prior to the sampling time. It was not possible to test such a dependence using the Perch Lake data since no water measurements were made prior to the May sampling period. Instead, most modelers based their OBT prediction on the predicted HTO concentration in the algae. These predictions showed greater scatter than those for HTO (Figure 3). Two modelers (IFIN and GE) attempted to simulate the formation of OBT using dynamic models that took into account the growth rate and dry fraction of the algae and the time-dependent water concentration, and both were relatively successful. The other participants assumed the OBT concentration was proportional to the HTO concentration, with the proportionality constant F_D allowing for processes such as isotopic discrimination. Most modelers took a value of F_D from the literature, with the chosen values ranging from 0.5 to 0.8 (Table 7). The BioM model calculated a somewhat lower value of 0.41 but this and the model prediction itself applies to buried tritium rather than OBT. The variation in F_D, coupled with the variation in the predicted HTO concentration in the plants, resulted in OBT predictions that varied by more than a factor of 2. Each individual prediction was within a factor of 1.7 of the observation, although in one case (J) the difference between prediction and observation was significant even when uncertainties were taken into account. Very similar results were obtained for sites S1 and S2. The full Perch Lake data set implies that $F_D = 0.46 \pm 0.08$ for algae, in agreement with the results of Blaylock et al. (1986).

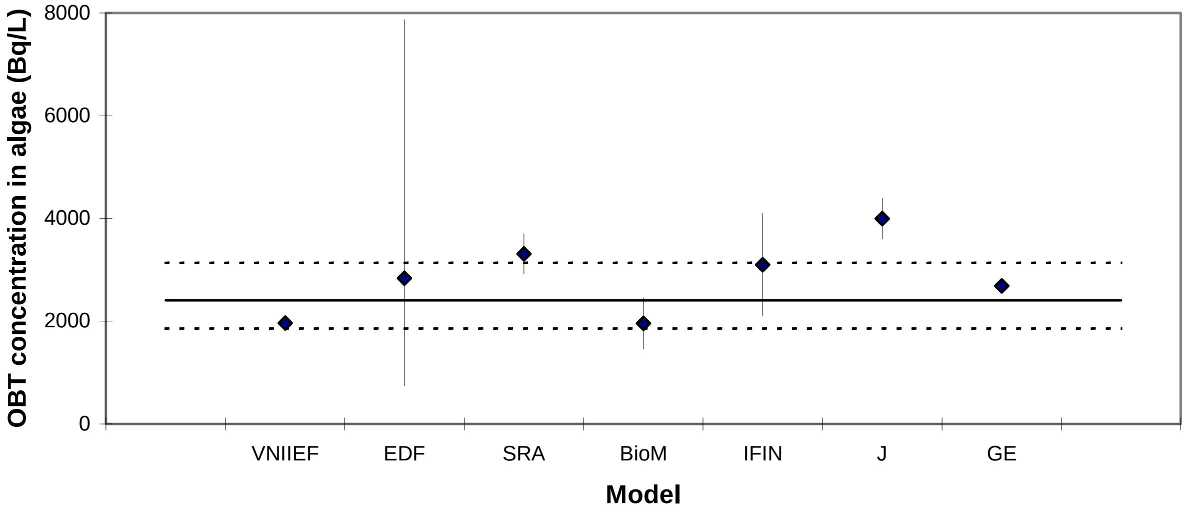

Figure 3. OBT concentrations in algae at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates and L did not submit results for this endpoint. The observed HTO concentration in algae at S3 in May was 4990 Bq/L.

Table 7. OBT/HTO ratios for plants

Participant	F_D = OBT/HTO			
	Algae	Worts	Cattails	
			Submerged	Emergent
VNIIEF	0.65	0.8	0.8	0.8
EDF	0.6	0.6	1.0	1.0
SRA	0.7	0.7	‡	0.7
BioM*	0.41	0.33	‡	0.33
IFIN	0.66[¶]	0.8	0.8	0.8
J	0.8	0.8	‡	0.8
GE	0.5[#]	-	-	-
L	-	0.75	0.82	0.66
Observed	0.46	0.48	0.70[†]	0.70

[*] Calculated for buried tritium from a two-step exchange process, taking into account the proportion of carbohydrates, proteins and DNA in the tissues

[¶] Calculated from a time-dependent model that depends on the algal growth rate and the HTO concentration in water

[#] Calculated from a time-dependent model that depends on the rates of algal metabolism and catabolism

‡OBT concentration in submerged cattails assumed equal to concentration in emergent parts

[†] Ratio of OBT concentration in submerged cattails to HTO concentration in emergent parts

The 95% confidence intervals shown in Figures 2 and 3 vary greatly from model to model, reflecting the different approaches taken by the participants in estimating their uncertainties. The confidence interval on the OBT concentration for model J is clearly an underestimate since the prediction does not agree with the observation even when uncertainties are taken into account. On the other hand, the confidence interval estimated by EDF (which reflects the variability in the observed water concentrations over all time and space) is so large that the prediction loses a lot of its usefulness. Similar variability arose for the other endpoints and will be discussed further in Section 5.

4.3 Worts: Predictions for the HTO concentration in worts at sampling site S3 in May are compared with the observation in Figure 4. As was the case for algae, the HTO concentration in worts collected at a given time and place is expected to equal the local water concentration, and all modelers made this assumption. Unfortunately, the worts were collected near shore in May and the water samples were taken off shore, so a local water concentration was not available. The participants approximated the missing data in various ways. SRA adopted the near shore water concentration observed in July at S3, J took the May offshore value at S3, and EDF reduced the observed offshore value at S3 in May by the ratio of near shore to offshore concentrations at S3 in July. As a result, the predictions for worts show greater scatter than for algae, but all lie within 50% of the observation and all agree with the observation when uncertainties are taken into account.

The results for S1 showed somewhat less scatter than for S3, but those for S2 showed greater scatter. At both S1 and S2, all of the predictions underestimated the observations. However, this may not be significant given the difficulty in estimating the water concentration at the location where the worts were sampled.

All modelers assumed that the OBT concentration in worts was proportional to the predicted HTO concentration. The OBT predictions showed greater scatter than those for HTO, ranging over a factor of 4 (Figure 5). This scatter was due to the variability in both the predicted HTO concentrations and the values chosen for the proportionality constant F_D, which ranged from 0.33 to 0.8 (Table 7). Only three of the results agree with the observation when uncertainties are taken into account. All but one of the predictions overestimate the observation, but this may be the fault of the observation, which appears low in relation to the measured HTO concentration in the plants. Similar results were obtained for sites S1 and S2, although here the model predictions scatter more uniformly about the observations. The full Perch Lake data set implies that $F_D = 0.48 \pm 0.19$ for worts.

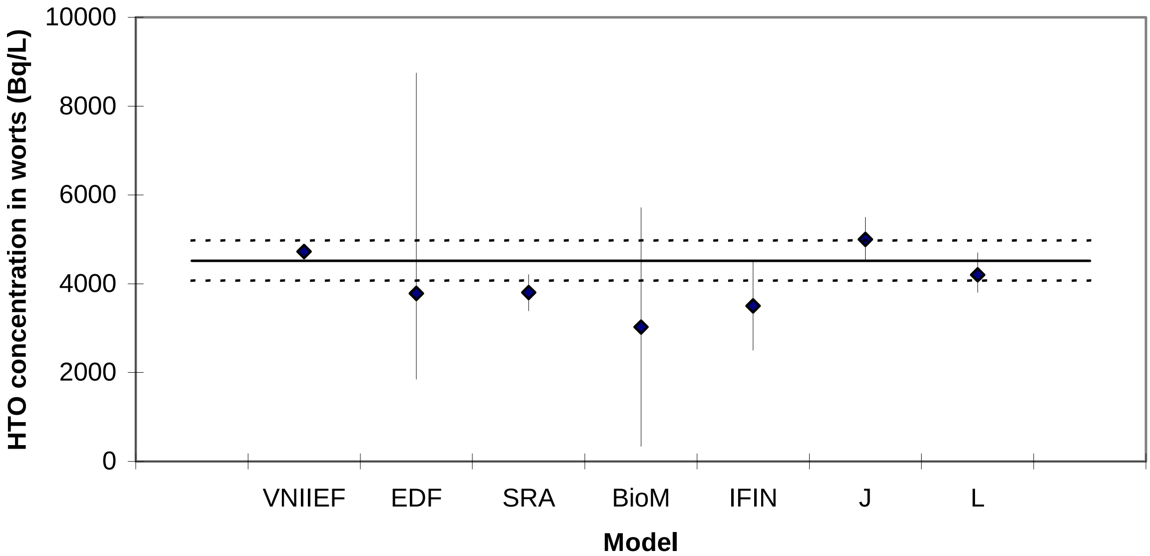

Figure 4. HTO concentrations in worts at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint. The HTO concentration in near-shore water at S3 in May was not measured.

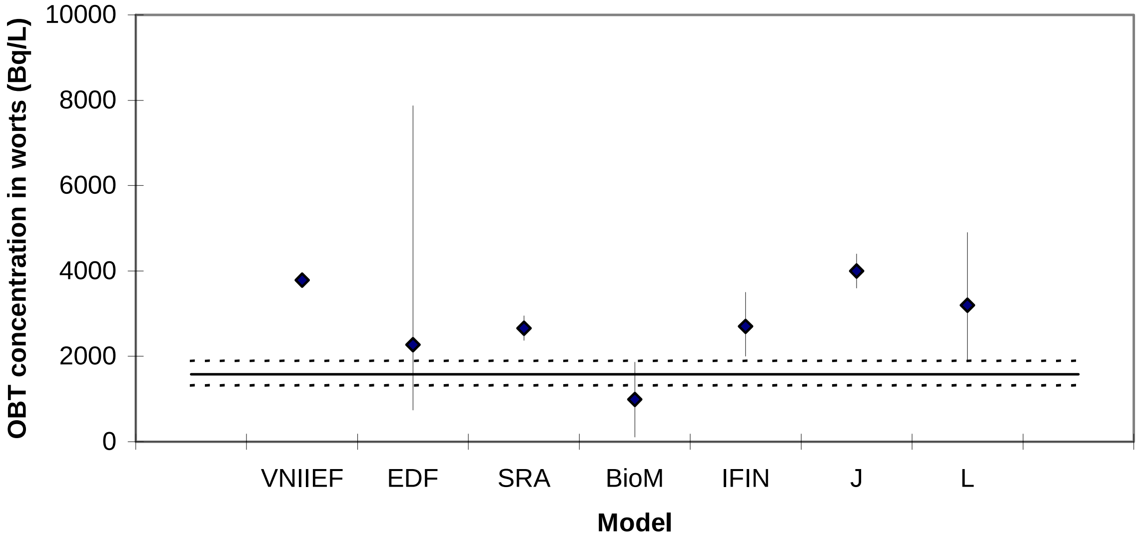

Figure 5. OBT concentrations in worts at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint. The observed HTO concentration in worts at S3 in May was 4520 Bq/L.

4.4 Cattails

4.4.1 HTO Concentrations in Submerged Cattails: Predictions for the HTO concentration in the below-water parts of the cattails at sampling site S3 in May are compared with the observation in Figure 6. Because cattails are rooted in the sediments, their HTO concentrations are expected to equal the concentration in sediment water. This approach was taken by three modelers (VNIIEF, SRA and BioM). Since the near-shore sediment concentration was not measured in May, two of the modelers (VNIIEF and BioM) used the off-shore value instead and obtained a result in close agreement with the observation. The third modeler (SRA) used the near-shore value for July and underpredicted severely. Four participants (EDF, IFIN, J and L) modeled the cattails in the same way as worts, setting the HTO concentration equal to the local lake water concentration. This approach overestimated the observation in each case, with none of the predictions agreeing with the observation even when uncertainties were taken into account.

The predictions for sampling site S1 all lay in the range 2000 Bq/L to 4000 Bq/L and all agreed reasonably well with the observation (3390 Bq/L). The good performance here is due to the fact that the sediment and water concentrations were similar and roughly constant in May and July. The scatter in the predictions for site S2 was about the same as for S3, ranging over a factor of 3.5. Here the models that were based on the water concentration underpredicted the observation by a factor of about 2.5, since the water concentration was less than the sediment concentration.

In contrast, the models based on sediment concentration returned predictions within 50% of the observation.

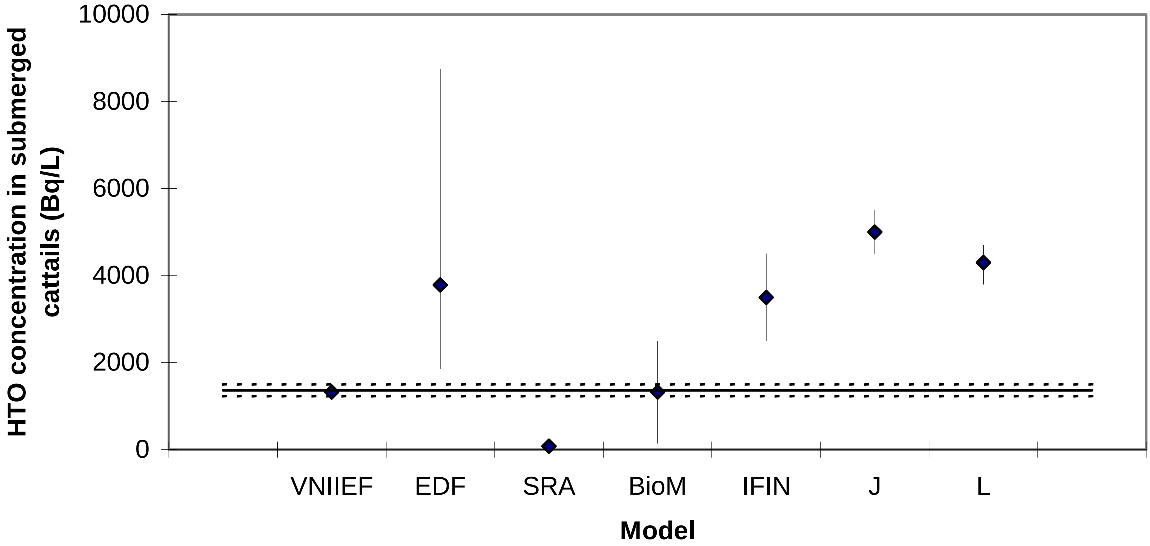

Figure 6. HTO concentrations in submerged cattails at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint.

4.4.2 HTO Concentrations in Emergent Cattails: Predictions for the HTO concentration in the emergent parts of the cattails sampled at site S3 in May are compared with the observation in Figure 7. As noted in Section 2.2, cattail concentrations are well predicted by the average of the concentrations in sediment water and air moisture. Three modelers (VNIIEF, EDF and SRA) explicitly took the contribution from air moisture into account. EDF took an average of the air and surface water concentrations, but would have done better to average air and sediment water. SRA assumed the cattail concentration was made up of 30% air and 70% sediment water (where the air concentration was set to 0) but underestimated severely because of an inappropriate choice for the sediment concentration. VNIIEF used a weighting of 75% air and 25% sediment water and produced a good result by using the offshore sediment water concentration measured at S3 in May. Participant J modeled emergent cattails in the same way as all other plants, setting the HTO concentration equal to the local water concentration, and overestimated the observation. IFIN and L lowered their predictions for cattails below those for other plants in recognition of the contribution from the air, but still overestimated the observation. The BioM result is also an overestimate since it predicts that the cattail concentration is slightly higher than the sediment concentration.

The predictions for sampling site S1 showed less scatter than those for S3 because of the similarity in the water and sediment concentrations at this site. The predictions ranged from about 1000 Bq/L to 4000 Bq/L compared to the observed value of 1970 Bq/L. The range in predictions for site S2 was larger (a factor of 7) because the sediment concentrations were more than twice the water concentrations. The models that were based on the water concentration underpredicted the observation whereas those based on sediment concentration overpredicted.

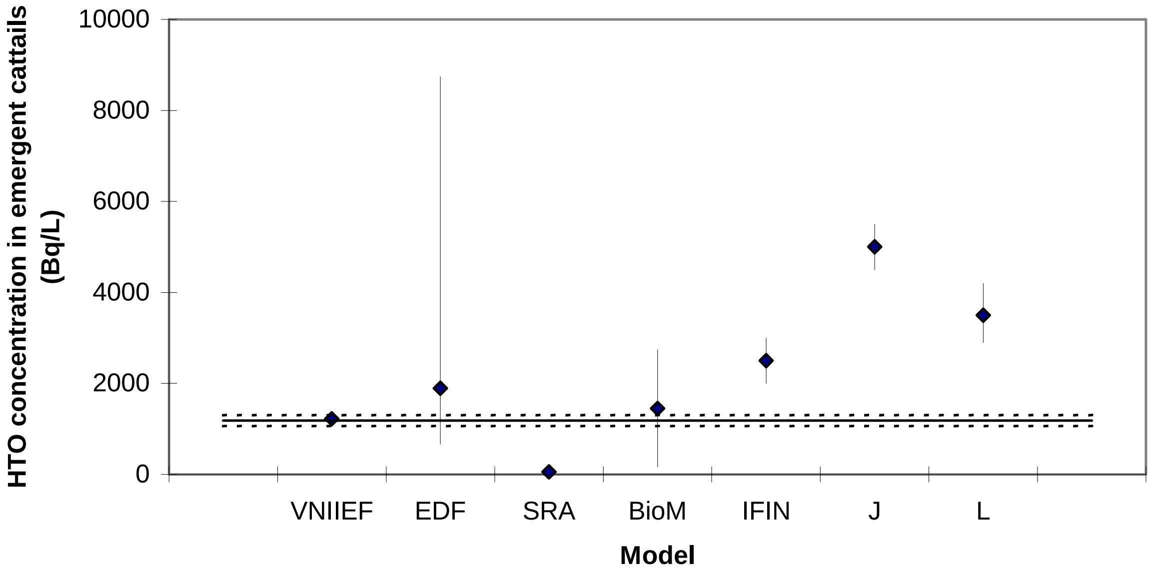

Figure 7. HTO concentrations in emergent cattails at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint.

4.4.3 OBT Concentrations in Cattails: Analysis of the full Perch Lake data set indicates that OBT concentrations are the same in both the emergent and submerged parts of the cattails, with a magnitude equal to 0.7 times the HTO concentration in the emergent part (Kim et al. 2004). This suggests that the OBT is formed primarily by photosynthesis in the emergent part and translocated to the submerged parts. Most modelers assumed that the OBT concentration in the emergent part was proportional to the HTO concentration in that part, with a proportionality constant F_D equal to that in the last column of Table 7. The results show considerable variability (Figure 8), due primarily to the differences among the predicted HTO concentrations, with some contribution from the values used for F_D. Only two predictions agree with the observation when uncertainties are taken into account. Three of the modelers (SRA, BioM and J) assumed that the OBT concentration in the underwater parts was the same as that in the emergent parts. Most of the other modelers calculated the OBT concentration in the submerged parts from the HTO concentration in the submerged parts, using the F_D values in the fourth column of Table 7. The comparison between predictions and observations for this endpoint (Figure 9) shows much the same pattern as for the emergent parts in Figure 8, with agreement in only three cases when uncertainties are taken into account. The results for sampling sites S1 and S2 are very similar to those for S3.

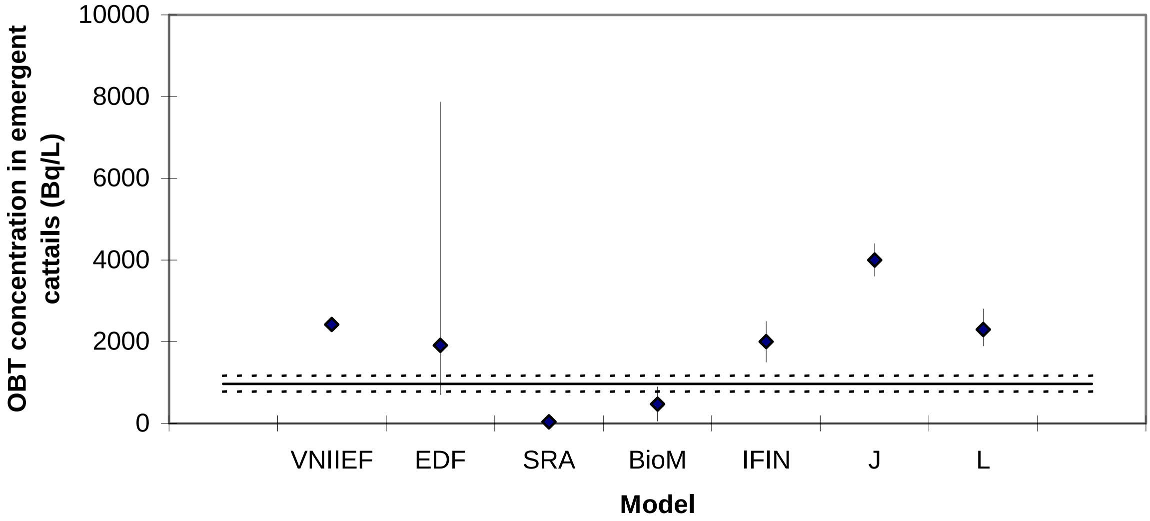

Figure 8. OBT concentrations in emergent cattails at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint.

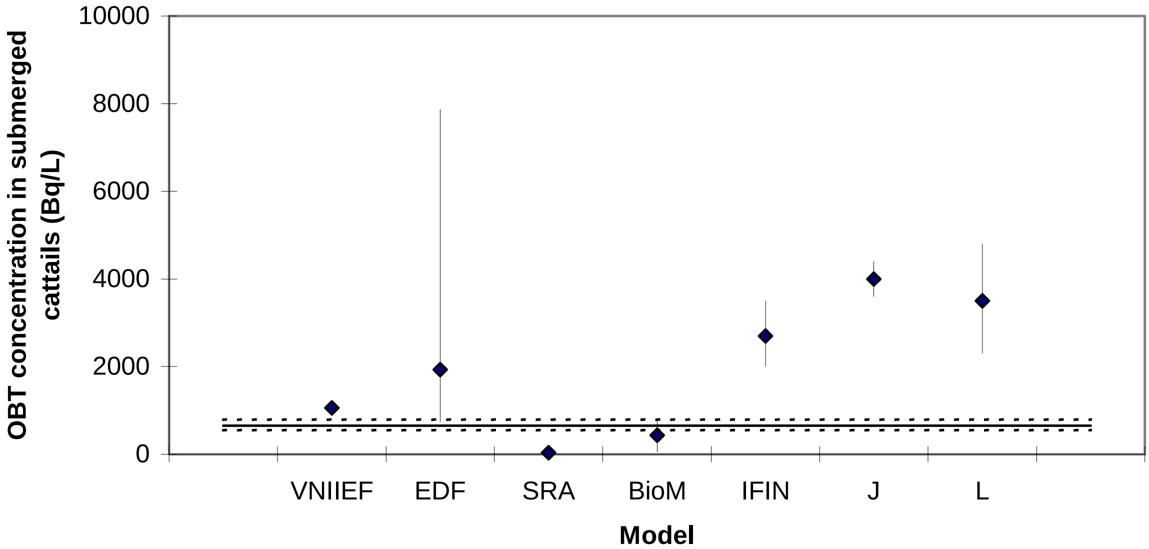

Figure 9. OBT concentrations in submerged cattails at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF did not provide uncertainty estimates and GE did not submit results for this endpoint.

4.5 Clams: Predictions for the HTO concentration in clams in July are compared with the observation in Figure 10. Because clams live at the sediment/water interface, their HTO concentration is expected to equal the local bottom water concentration at the time of sampling. Most participants made this assumption but, in the absence of measured water or sediment concentrations in the area where the clams were harvested, they estimated the water concentrations in different ways. In the case of EDF, the concentrations were calculated as the average of the near shore and offshore sediment concentrations for the three sampling sites; for SRA, as the average of the deep and surface water concentrations at S1 and S3 and the sediment water concentration at S3; and for IFIN, as the average of the bottom water and sediment concentrations over time throughout the lake. Despite these different approaches, the predictions agreed with the observation for each model in which uncertainties were estimated (Figure 10), and five of the eight predictions lay within 12% of the observation. Similar agreement was obtained for the May sampling period. The uncertainties were large for some models, reflecting the difficulties the modelers had in estimating the water concentrations experienced by the clams. Analysis of the full Perch Lake data set (Kim et al. 2004) indicates that the clam concentration in July (4100 Bq/L) lay within 10% of the average offshore bottom water concentration (4495 Bq/L).

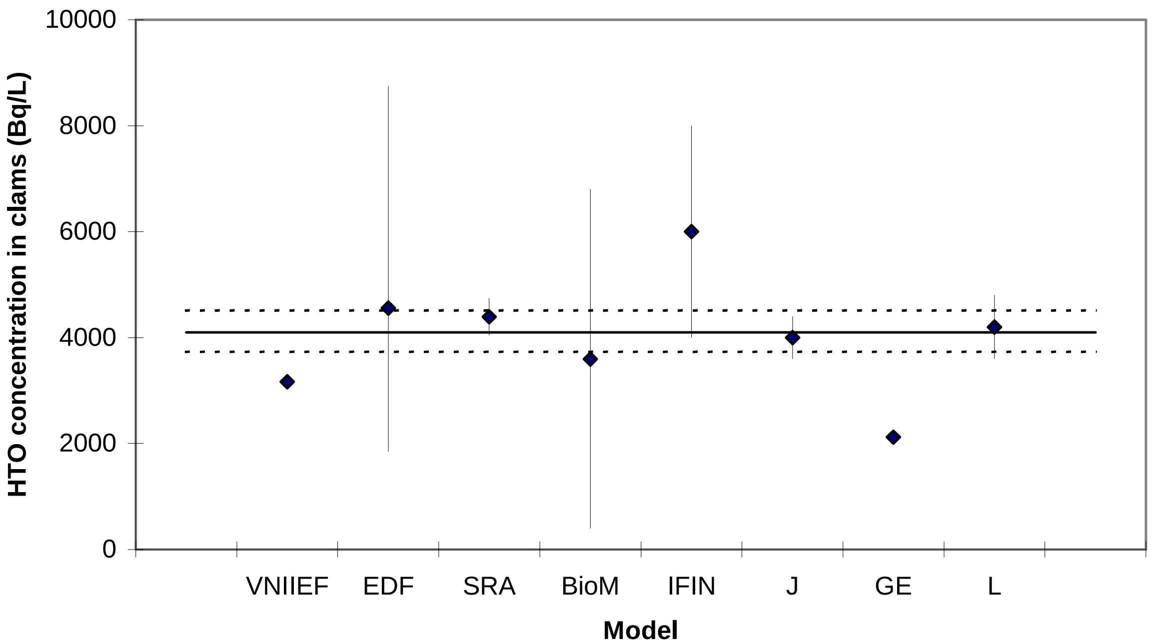

Figure 10. HTO concentrations in clams in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates for this endpoint.

Clams are filter feeders, eating phytoplankton and zooplankton but also retaining detritus. Most OBT in clams and other aquatic animals is the result of direct incorporation of OBT taken in with the diet. However, only two of the participants (IFIN and GE) simulated OBT formation in this way, using dynamic models that took into account the metabolism/catabolism of the animals and the time-dependent water concentrations. IFIN overestimated the observation for clams by about 50% whereas GE underestimated by about 20% (Figure 11). The latter model predicted an OBT/HTO ratio of 1.5, implying bioaccumulation of tritium in the organic material of the clams. Most of the remaining participants assumed the OBT concentration was proportional to the HTO concentration in the clams, with the proportionality constant, F_M, accounting for metabolic processes. Most modelers took a value of F_M from the literature, with the chosen values ranging from 0.30 to 0.95 (Table 8). In most cases, the HTO concentrations used were those predicted for July, even though the slow turnover rate of OBT in animals implies that they should be based on HTO concentrations integrated over the few weeks prior to sampling. The predictions showed greater scatter than those for HTO (Figure 11). The variations in F_M, coupled with the variations in the predicted HTO concentration in the clams, resulted in OBT predictions that varied by more than a factor of 5. Seven of the eight models underpredict and only two of the predictions agree with the observation even when uncertainties are taken into account. Despite the added complexity in predicting OBT, the uncertainties assigned to several of the OBT predictions were smaller than those for the corresponding HTO concentrations.

Table 8. OBT/HTO ratios for aquatic animals

Participant	F_M = OBT/HTO						
	Clams	Bullheads			Pike		
		Flesh	Head	Organs	Flesh	Head	Organs
VNIIEF	0.75-0.95	0.4	0.3	0.5	0.27	0.3	0.2
EDF§	0.45	0.52 – 0.57 depending on the month					
SRA	0.5	0.7	0.64	0.66	0.7	0.64	0.66
BioM*	0.30	0.3	0.1 for gonads		0.3	0.1 for gonads	
IFIN†	0.92	0.93-1.2	Higher for viscera		1.0	Higher for viscera	
J	0.8	0.8	0.8	0.8	0.8	0.8	0.8
GE‡	1.48	1.48 – 2.05 depending on the month					
L	0.38	0.38	0.38	0.38	0.38	0.38	0.38
Observed	0.75	0.77			0.84		

§ Animal OBT/water HTO = 0.45 for clams and all parts of fish

* Calculated from a two-step exchange process for buried tritium, taking into account the proportion of carbohydrates, proteins and DNA in the tissues

† Calculated from a time-dependent model that depends on mass, metabolic rate and OBT residence time

‡ Calculated from a time-dependent model that depends on the rates of metabolism, catabolism, ingestion and excretion

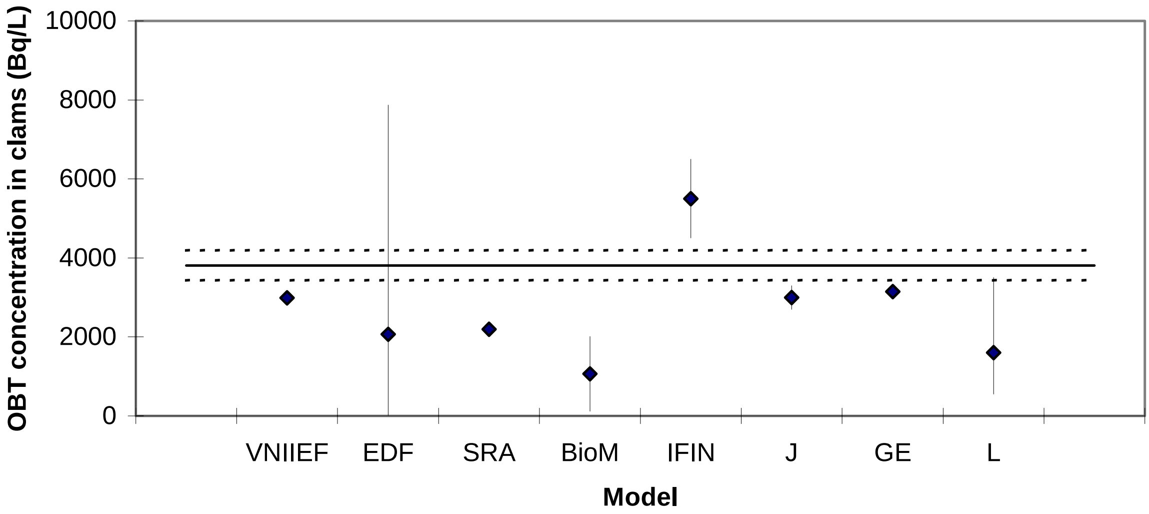

Figure 11. OBT concentrations in clams in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates for this endpoint.

The results for May showed more scatter but less bias than those for July, with half of the models overestimating the observation and half underestimating. Based on an analysis of the full data set, Kim et al. (2004) found good agreement between predictions and observations when the OBT concentrations in clams were calculated by multiplying the HTO concentration in bottom water (averaged over the entire lake and over time up to the time of sampling) by a metabolic factor $F_M = 0.75$.

4.6 Bullheads: Bullheads are benthic fish that move freely throughout the lake near the sediment/water interface. They are omnivorous, eating a variety of molluscs, insects, leeches, worms, algae, plant material and small fish. Because of the rapid rate of equilibrium between HTO in lake water and fish, the HTO concentration in bullheads is expected to equal the average concentration in the water encountered by the fish in the hour or two prior to sampling. The analysis of the full Perch Lake dataset showed that the observed HTO concentrations in bullheads were essentially equal to the concentration in bottom waters averaged over the entire lake at the time the fish were sampled. EDF, BioM, IFIN and L all based their predictions on the average HTO concentration in offshore waters only and slightly overestimated the observation (Figure 12), since offshore waters had a higher concentration than near-shore waters. The high result of SRA is due to the fact that, in this model, half of the tritium in the fish was assumed to come from sediment waters, which had high concentrations at some times and locations in the lake. The other participants adopted a water concentration lower than the average observed concentration for bottom waters and underestimated the observation. The predictions of SRA and J do not agree with the observations even when uncertainties are taken into account. The predictions for May and October show more scatter than for July, likely because the HTO concentrations in the environment were less well characterized and it was more difficult to define a representative water concentration for the bullheads.

All models but one predicted equal HTO concentrations in all parts of the fish, in agreement with the observations. The exception was VNIIEF, where the concentrations in internal organs were sometimes higher and sometimes lower than in the flesh and head. In this model, HTO in the head is assumed to come from the water column and HTO in the organs from the diet of the fish; HTO in the flesh comes partly from the water and partly from the diet.

Predicted and observed OBT concentrations in bullhead flesh in July are shown in Figure 13. Two of the process-oriented models (IFIN and GE) substantially overestimated the observation and both predicted OBT/HTO ratios in the fish greater than one. The other dynamic model (EDF) slightly underestimated the observation. As was the case for clams, most of the remaining participants assumed the OBT concentration was proportional to the HTO concentration, with the proportionality constant F_M shown in Table 8. Kim et al. (2004) found good agreement between predictions and observations when the OBT concentrations in bullheads were calculated from the HTO concentration in bottom water averaged over the entire lake and over time up to the time of sampling, with $F_M = 0.77$. Most modelers used a lower value, which explains in part why most predictions underestimate the OBT concentration in bullheads. The differences in model formulation and parameter values adopted by the various participants resulted in OBT predictions that ranged over more than a factor of 6. Individual predictions differed from the observation by up to a factor of 4, and in only three cases did the prediction and observation agree when uncertainties were taken into account. Very similar results were obtained for the May and October sampling periods.

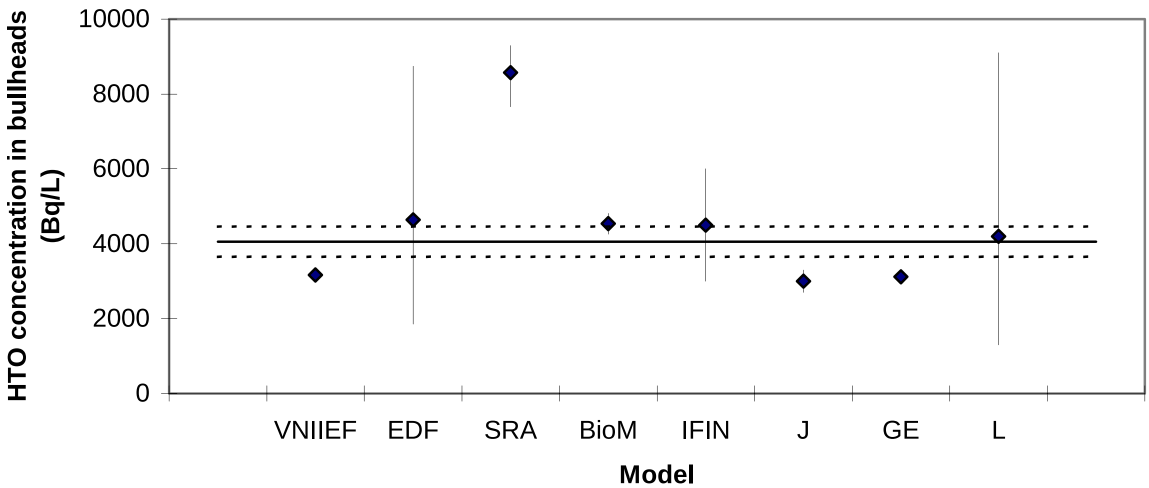

Figure 12. HTO concentrations in bullheads in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates for this endpoint. The observed HTO concentration in bottom waters averaged over the entire lake for July was 4000 Bq/L.

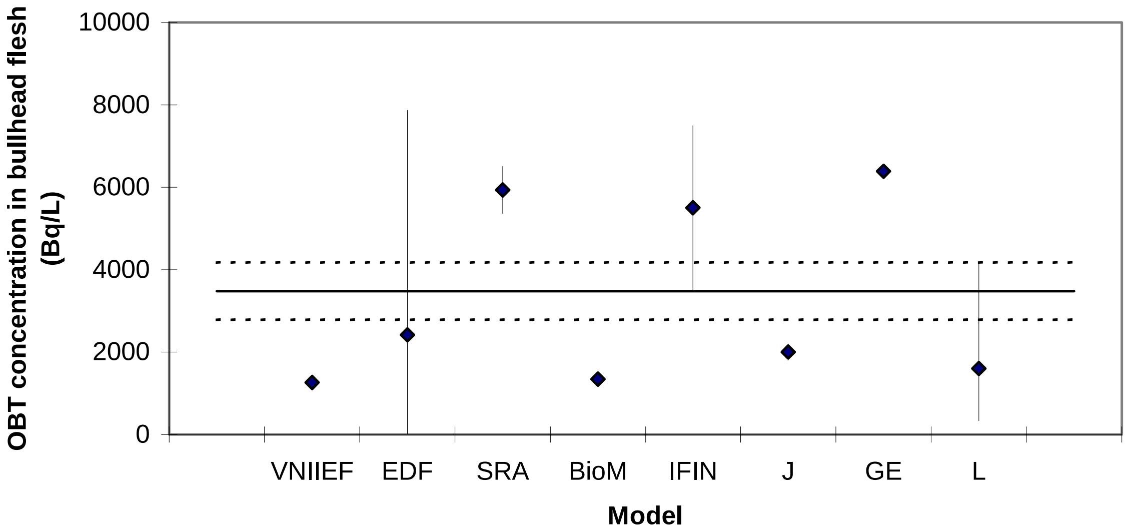

Figure 13. OBT concentrations in bullhead flesh in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not submit uncertainty estimates for this endpoint. The observed HTO concentration in bottom waters averaged over the entire lake over the May and July sampling periods was 4655 Bq/L.

Half of the models (VNIIEF, SRA, BioM and IFIN)) showed different OBT concentrations in the different parts of the fish, reflecting the different proportions of proteins, carbohydrates and fat in the flesh, head and internal organs. The differences were small for SRA (10%) and IFIN but more substantial for VNIIEF (67%) and BioM (a factor of 3 between flesh and gonads, but this result applies to buried tritium rather than OBT). In contrast, the data show that OBT concentrations in the flesh, head and internal organs of the bullheads are not significantly different, an assumption made by models EDF, J, GE and L.

4.7 Pike: Five participants (EDF, BioM, J, GE and L) modeled pike in the same way as bullheads and predicted the same tritium concentrations for both types of fish. These modelers felt either that the foraging habits of bullheads and pike were sufficiently similar that they could be modeled in the same way, that there was too little information to attempt to model them differently, or that any differences in habits would not translate into significant differences in concentration in a well-mixed system such as Perch Lake. For the IFIN model, results for pike and bullheads differed by less than 10%, whereas the results for VNIIEF were within 45%. SRA predicted concentrations in pike that were a factor of 2 lower than those in bullheads on the assumption that sediment water plays less of a role in determining tritium levels in pike than in bullheads. In fact, the experimental data indicate that concentrations in the two types of fish are identical within measurement error. As piscivores that move freely throughout the lake, pike differ from bullheads in the parts of the lake they access and the type of food they eat. However these differences in behaviour do not result in significant differences in concentrations in Perch Lake.

The observed and predicted HTO concentrations in pike in July are shown in Figure 14. The overall results are good and only two predictions (those for J and GE) do not agree with the observation when uncertainties are taken into account. The predictions for VNIIEF and SRA are much better for pike than they were for bullheads. In contrast, most of the models underestimate the observed OBT concentration in pike flesh for July (Figure 15) because they underestimate the metabolic factor F_M. Similar results were obtained for the May and October sampling periods.

As was the case for bullheads, the observed tritium concentrations were the same to within measurement error in all parts of the pike. Of all the predictions, only those of VNIIEF and BioM are inconsistent with this finding.

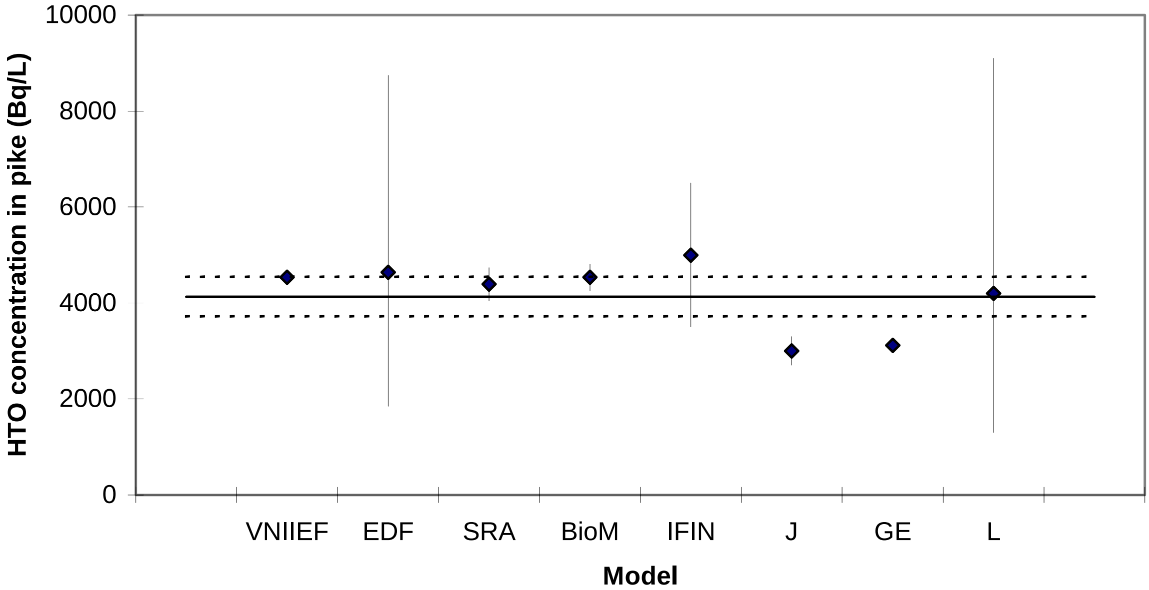

Figure 14. HTO concentrations in pike in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates for this endpoint. The observed HTO concentration averaged over the entire lake for July was 4130 Bq/L.

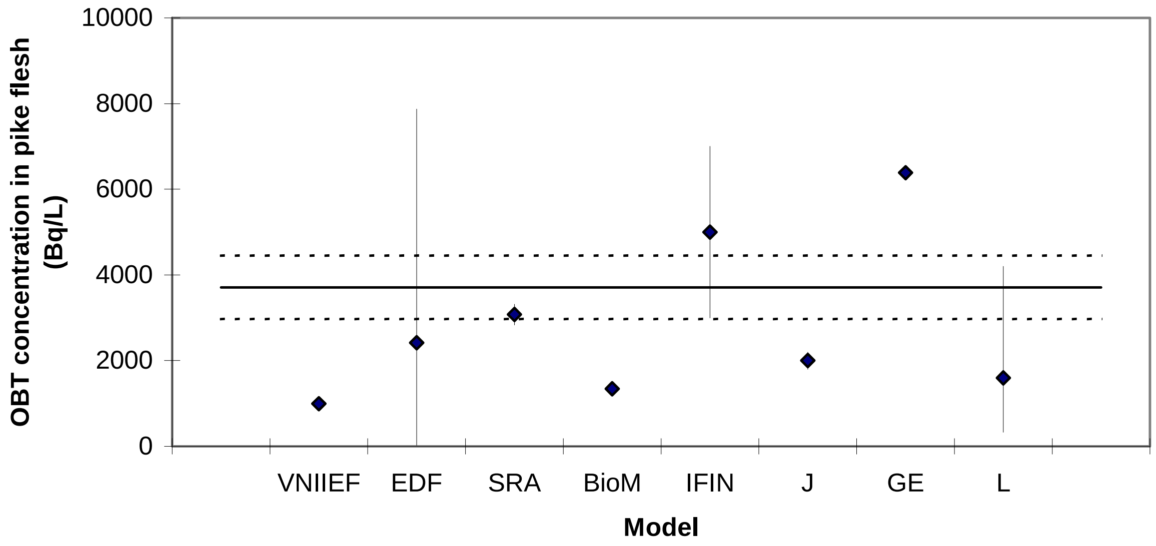

Figure 15. OBT concentrations in pike flesh in July. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. VNIIEF and GE did not provide uncertainty estimates for this endpoint. The observed HTO concentration averaged over the entire lake over the May and July sampling periods was 4720 Bq/L.

4.8 Sediments: There is no evidence that OBT discharges directly to the lake with groundwater. If this is the case, sediment OBT must arise from decaying plant and animal material deposited on the lake bottom, with the vast majority expected to come from plants. The experimental data suggest that the mean sediment/plant ratio is 0.61 ± 0.20. Since most sediments were collected from shore, only those plants found close to shore (worts and cattails) were considered in this calculation. Also, the plant concentrations were averaged over time up to the time of sampling, to account in a small way for the fact that the sediments, which were collected to a depth of 15 cm, are averages of the material deposited over a considerable length of time. The sediment concentrations are believed to be lower than those in plants due to radioactive decay and/or the breakdown over time of OBT in the decomposing plant material.

Predicted and observed sediment OBT concentrations at sampling site S3 in May are shown in Figure 16. The predictions range over a factor of 100 and only two of the six predictions agree with the observation when uncertainties are taken into account. The variation is due to the very different assumptions made by each modeler in calculating the sediment concentrations:

- VNIIEF: the sediment OBT concentration was assumed equal to the HTO concentration in detritus formed in surface waters in May.
- EDF: the sediment OBT concentration at S3 was assumed to be in equilibrium with the OBT concentration in the organic matter of decomposing terrestrial vegetation, which was assumed to equal 60% of the air HTO concentration.

- SRA: the sediment OBT concentration was assumed equal to 0.63 times the HTO concentration in the near-shore sediment water.
- BioM: the concentration of buried tritium in sediment was assumed to equal the predicted concentration of buried tritium in the submerged part of cattails.
- IFIN: the sediment OBT concentration was estimated from the predicted OBT concentration in macrophytes and benthic algae, which in turn depend on the HTO concentration in bottom waters.
- J: the sediment OBT concentration was set equal to the mean of the predicted plant and animal OBT concentrations.

Most of these assumptions were reasonable, but only BioM produced a result in good agreement with the observation. This must be considered fortuitous since BioM predicts the concentration of buried tritium whereas the observation is of organically bound tritium. The other models did not do as well because they all overestimated the concentrations in the plants that were assumed to make up the sediments.

The results for sites S1 and S2 showed somewhat less scatter than for S3, although the predictions still ranged over a factor of 3 or 4, and most of the models continued to overestimate the sediment concentrations.

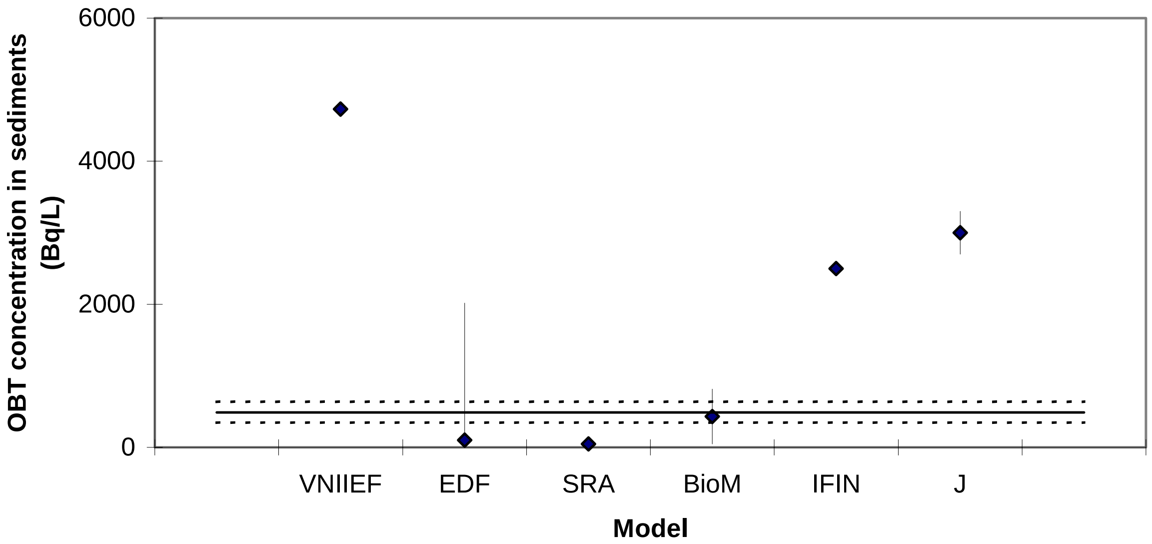

Figure 16. OBT concentrations in sediments at site S3 in May. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. GE and L did not submit predictions for this endpoint. The observed OBT concentration in worts and cattails at S3 in May was 1070 Bq/L.

5. DISCUSSION AND CONCLUSIONS

The Perch Lake scenario provided a good test of models that predict tritium concentrations in the various compartments of a freshwater ecosystem at steady state. Apart from a narrow zone close to shore near the inlets, the lake is well mixed with respect to HTO concentrations in the water, and concentrations change only slowly over time. Therefore the water concentrations to which the fish are exposed, and the concentrations in the plants and animals that make up their diets, can be estimated with some confidence. Moreover, the concentrations in sediments are substantially different from those in the lake water itself, which makes it possible to say whether the tritium in plants and fish came from the water or the sediments. On the other hand, the scenario was not ideal since some relevant information was missing or incomplete, and this contributed to the differences between predictions and observations. But many real assessments must be carried out with even less information, and discrepancies of a similar magnitude must be expected in practice.

A number of conclusions regarding the relationship between tritium concentrations in the various parts of the Perch Lake ecosystem can be drawn from an analysis of the full data set (Kim et al. 2004) and the results discussed here:

- The HTO concentration in a given plant or animal is equal to the concentration in water, sediments or air to which the organism was exposed in the hour or two prior to sampling. For algae and worts, this is the local concentration in water. For submerged cattails it is the sediment water concentration and for emergent cattails, an average of air and sediment concentrations. Concentrations in clams and bullheads are the same as the concentrations in local bottom waters, and bottom waters averaged over the entire lake, respectively. HTO concentrations in pike reflect an average of both bottom and surface waters over the entire lake.

- The OBT concentration in algae and worts is about half the HTO concentration in the plant. The OBT concentration in the emergent parts of cattails is about 70% of the HTO concentration. The OBT concentration in the submerged parts of cattails is the same as in the upper part, indicating that the OBT forms in the emergent parts and is translocated to the parts below water.

- The OBT concentration in clams, bullheads and pike is about 80% of the HTO concentration in the water to which the animal is exposed.

- The OBT concentration in sediments is about 60% of the OBT concentration in the aquatic plants that make up most of the organic fraction of the sediments. The sediment concentrations are believed to be lower than those in plants because of radioactive decay and/or the breakdown over time of OBT in the decaying plant material.

- The OBT concentration in each compartment should be calculated from the HTO concentration averaged over the few weeks prior to sampling.

- Within measurement error, there is no significant difference between the HTO or OBT concentrations in different parts of the fish.

When the predictions of all the models were averaged for a given endpoint, the mean lay within 30% of the observation in each case except for OBT concentrations in sediments and the underwater parts of cattails. With these exceptions, the modelers as a group have a good conceptual understanding of the behaviour of tritium in the Perch Lake ecosystem and can predict HTO and OBT concentrations that, in an average sense, agree well with the observations. However, the difference between prediction and observation for an individual model could be as large as a factor of 25. More typically, the predictions of a given model for HTO concentrations in plants and animals lay within 30% of the corresponding observation, and the predictions of OBT concentrations within a factor of about 2. These differences for OBT are significant even when uncertainties are taken into account. The models were equally as likely to overpredict as to underpredict the HTO concentrations in plants. They tended to be conservative for HTO concentrations in animals and OBT concentrations in plants but to underestimate OBT concentrations in animals.

There were several reasons for the mispredictions:

- An inappropriate choice for the source compartment from which the plant or animal draws its tritium. In particular, the submerged parts of cattails are in equilibrium with sediment water rather than lake water; clams and bullheads are in equilibrium with bottom water rather than sediment water; and OBT in submerged cattails is translocated from the emergent parts of the plant rather than being formed in place.

- An inappropriate choice of surrogate values when HTO concentrations in the source compartment were not available. The modelers had particular difficulty in defining the source terms for worts and cattails, since near-shore water and sediment concentrations were not measured in May. Similarly, no sediment or water concentrations were measured in the area where the clams were harvested.

- An inappropriate choice for the discrimination and metabolic factors, F_D and F_M, used to calculate OBT concentrations from the HTO concentrations.

- Inappropriate spatial averaging, particularly for fish. The best prediction of HTO concentration in bullheads was obtained by averaging the bottom waters over the entire lake, including near-shore and offshore zones. Similarly, the best prediction of HTO concentration in pike occurs by averaging over the water column as well as over the entire lake.

- Lack of time-averaging when calculating OBT concentrations. Apart from the dynamic results for algae, clams and fish generated by IFIN and GE, none of the models considered any sort of time-averaging in calculating OBT concentrations in plants or animals. In contrast, the observed OBT concentrations correlate better with the time-averaged HTO concentrations than with point concentrations.

No one model stood out as generating predictions superior to the others. The level of agreement between predictions and observations was about the same for the dynamic models as for the steady-state models, although the dynamic models tended to have the highest predictions for OBT concentrations in clams, bullheads and pike. None of the models were satisfactory for sediments.

The results of the BioM model, which calculates the concentration of buried tritium rather than the tritium traditionally considered to be organically bound, were generally lower than those of the other models for the OBT endpoints. However, the BioM predictions made up a substantial proportion (between 25% and 90% depending on the endpoint) of the measured OBT concentrations. If the results of this model are correct, this implies that the fraction of carbon bound tritium in the OBT yielded by conventional analytical techniques is much lower than normally believed. This could have consequences for dose estimation, although such consequences may be small since the dose conversion factors for OBT are based on OBT concentrations measured in the traditional way. The results of the BioM model indicate that the formation of buried tritium is better modeled as a two-step exchange process rather than as a one-step process.

Despite that fact that two models predicted OBT/HTO ratios greater than one for some endpoints, there is no evidence in the Perch Lake data of tritium bioaccumulation in OBT formation. Ratios greater than one are confined to non-equilibrium situations such as those that exist in Cardiff Bay, where tritiated organic material is released directly to the water body (Williams et al. 2001, Lambert 2001).

Given the large variation in the confidence intervals estimated by the various participants, no definitive conclusions can be drawn regarding the uncertainties in the model predictions. Ideally, the confidence intervals would take into account the uncertainties in the HTO concentrations in water, sediments and air used to drive the models; in the conceptual models themselves to cover uncertainties in the appropriate source compartments and spatial and temporal averaging; in estimating values to replace missing data; and (for OBT concentrations only) in the parameters F_D and F_M. The dynamic models used by some participants would have additional sources of uncertainty associated with the extra parameters that are needed to describe the growth of the organisms and the metabolic processes that occur in them. The uncertainties are limited to some extent by specific activity concepts, since concentrations in a given compartment cannot be higher than concentrations in a donor compartment. Table 9 lists approximate 95% confidence intervals for the various endpoints based on an overall assessment of the differences between the observations and the predictions submitted by the participants. Hopefully the lessons learned in this scenario will help to reduce the uncertainties in future studies that require the estimation of steady-state tritium concentrations in freshwater ecosystems.

Table 9. 95% confidence intervals based on the differences between predictions and observations

Endpoint	95% confidence interval
HTO in algae, worts and all animals	BE* ± 30%
HTO in cattails	BE/2 to 2 BE
OBT in algae, worts	BE/2 to 2 BE
OBT in cattails	BE/3 to 3 BE
OBT in animals	BE/2.5 to 2.5 BE
OBT in sediments	BE/10 to 10 BE

* best estimate

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APPENDIX A

Perch Lake Scenario Description – Revision 1
EMRAS Tritium/C14 Working Group
January 2004

BACKGROUND

Located on the site of Chalk River Laboratories (CRL), Perch Lake contains trace amounts of tritium due to leakage from a nearby waste management area. The releases have been going on for many years and concentrations in various parts of the lake ecosystem are likely to be in equilibrium. Tritium concentrations in lake water, sediments, aquatic plants, fish, clams and air were collected three times during the summer and fall of 2003 at three locations in the lake. These data are offered here as a test of models that predict the long-term average tritium concentrations in aquatic systems due to chronic releases.

SITE DESCRIPTION

Perch Lake (Figure 1) is a small, shallow freshwater Canadian Shield lake. Its largest fetch is about 800 m and it has a surface area of 4.5 x 10^5 m^2. It has a mean depth of 2.0 m, a maximum depth of 4.1 m and a total volume of 9.1 x 10^5 m^3. It drains a watershed of area 5.65 x 10^6 m^2. The lake can be considered unstratified, although there is weak stratification in deeper areas in the summer, when surface waters are approximately 5^{o}C higher than those at lake bottom. The lake is usually ice covered from early December to mid April. Mean monthly water temperatures are 13, 19, 24, 23, 19 and 11^o C for the months of May through October. The turnover time of lake waters is about two years.

Sediments in the lake are composed of sand and gyttja (decomposing organic material). The average dry bulk density is 185 kg m^{-3} but this varies substantially across the lake depending on the local composition of the sediments. The sediments near Inlet 1 are largely organic in composition, those near Inlets 2 and 3 contain more sand and those near Inlet 4 and the outlet are primarily sand. The sediments are 89% water by weight and the sedimentation rate is 0.16 kg m^{-2} a^{-1}, or 0.06 cm a^{-1}.

Perch Lake is contaminated by tritium migrating through an extensive sand aquifer from a waste management area (WMA) located about 750 m to the north. The WMA was in operation for about 40 years until it was shut down in 1999. The tritium forms a well-defined underground plume that is narrow near the source but broadens to a width of about 1000 m by the time it reaches the lake. Tritium in the form of HTO discharges into the lake through the sediments from below and also through a stream (Inlet 2 in Figure 1) that flows above the underground plume. Inlet 1 shows slightly elevated levels of tritium but Inlets 3, 4 and 5 are all uncontaminated. The rate and distribution of HTO releases to the lake are not known quantitatively.

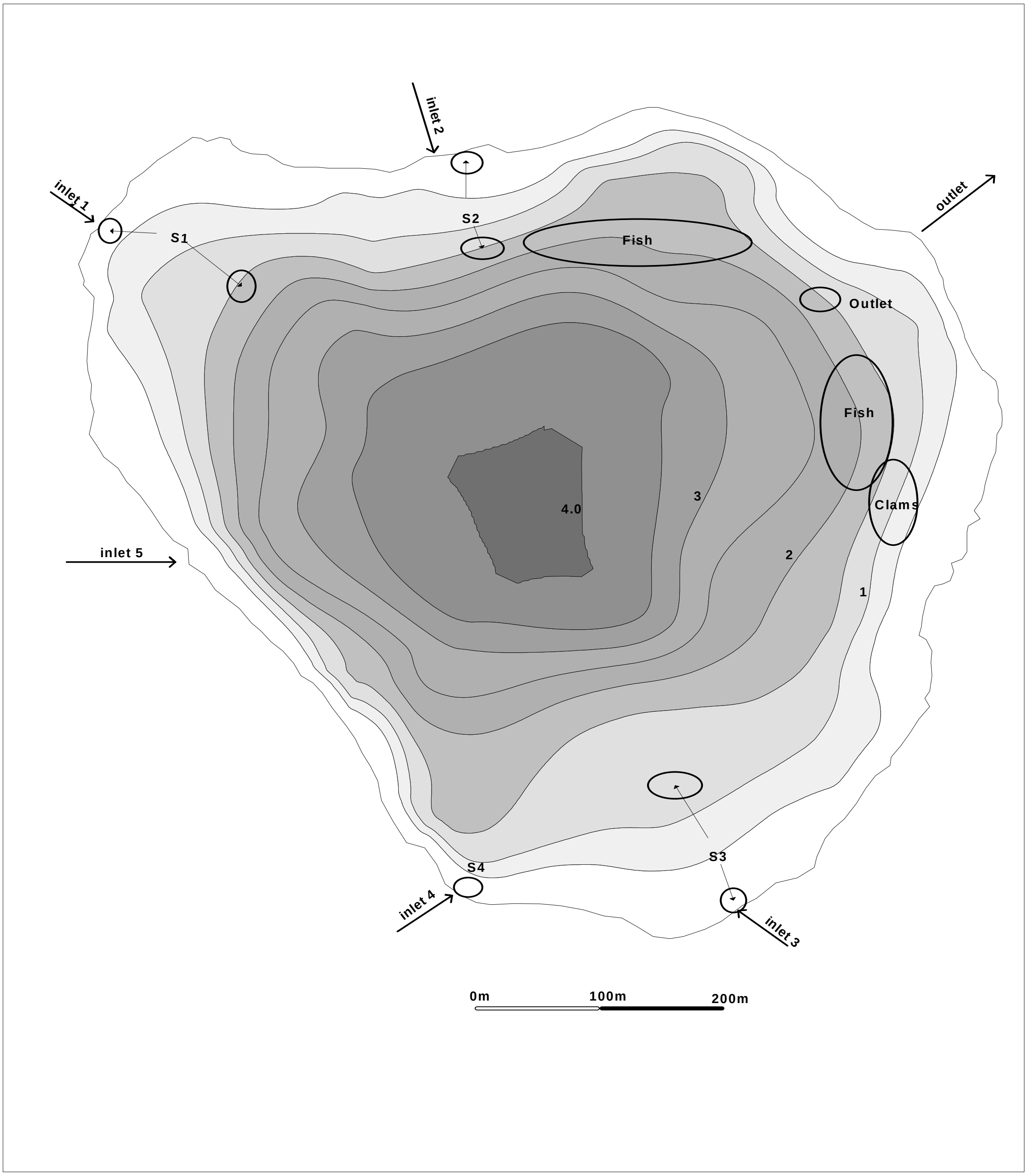

Figure 1. Map of Perch Lake showing inlets, the outlet, depth contours in m and the sampling locations.

TRITIUM MEASUREMENTS

Water, sediment, plant and air samples were collected primarily from three locations in Perch Lake: at S1, located near Inlet 1; at S2 near Inlet 2; and at S3 near Inlet 3 (Figure 1). A few samples were also taken at S4 near Inlet 4 and near the outlet of the lake. Most of the plant and sediment samples were collected from shore at the edge of the lake. Some of the water samples were also taken close to shore but others were collected by boat 50-100 m offshore, as were algae. Fish tend to feed on the east side of the lake and were caught in two extended areas on either side of the outlet, whereas clams were harvested between Inlet 3 and the outlet. Most samples were collected three times during the summer and fall of 2003 (May 27-28, July 28-29 and September 28-October 1). Additional measurements of water concentrations were made in early November. Air concentrations were measured only in August and September as monthly averages and algae were not available in September, as they had all died off. Replicate samples were taken in some cases.

Water: Water samples were collected near the surface of the lake and at deeper levels by opening sampling bottles at the desired depth. The samples were left standing to allow suspended sediments to settle out and then 10 ml of water was transferred to scintillation vials. HTO concentrations were determined by liquid scintillation counting (LSC).

Sediments: Sediment samples were scooped up by hand and placed in vinyl bags that were sealed at depth. This provided samples averaged over the top 15 cm or so of sediments. Water was extracted from the sediments by freeze-drying and analyzed for HTO concentration by LSC. The pressure during freeze-drying was between 10^{-4} and 10^{-5} Torr and the temperature was between 0 and -4^{o} C. The remaining solid material was washed with tritium-free water to remove the exchangeable OBT and was then completely dried in an oven, followed by combustion in a combustion tube. The combustion water was analyzed by LSC to give OBT concentrations.

Plants: Samples were taken of bladderwort (Utricularia spp.), hornwort (Ceratophylum demersum) and cattails (Typha latifolia), and of algae belonging to the phylum Chlorophyta. Bladderwort and hornwort are both unrooted plants that are completely submerged and obtain their nutrients from the water. They consist of a long thin stem that supports masses of delicate, needle-shaped, whirled leaves. These two species were composited for analysis. The cattails are rooted in the top 5-10 cm of the sediments, from which they draw their nutrients. They extend above the water into the air, and the submerged and emergent parts were analysed separately. Algae were scooped out of the water by hand and placed in a sampling jar after allowing the water to drain away. The bladderwort, hornwort and cattails were sampled from shore at the edge of the lake whereas the algae were collected further offshore. The water in all plant samples was extracted by freeze-drying and HTO concentrations were determined by LSC. The solid matter was washed with tritium-free water and was then oven-dried and combusted in a combustion bomb. LSC of the combustion water yielded non-exchangeable OBT concentrations.

Table 1 shows measured water contents of the aquatic plants. No data could be found on the nutrient composition (protein, fat and carbohydrate) of these plants.

Table 1. Plant water contents.

Plant type	Water Content (% by weight)
Algae	88.0
Bladderwort, hornwort	95.0
Cattail - below water	93.5
- above water	85.1

Aquatic Animals: The aquatic animals collected included clams (Elliptio complanata), bullheads (Ameiurus nebulosus) and pike (Esox lucius). Bullheads are small benthic fish and pike are larger piscivores. Both types of fish likely move throughout the lake, eating other fish and invertebrates. The fish were caught in nets and the clams were pulled individually from the sediments by hand. The fish samples were divided into three parts (flesh, head and internal organs), each of which was analyzed separately. About five pike, 20 bullheads and 12 clams were combined to provide enough mass for each analysis. The fish caught in May were significantly smaller (mean weight 40 g for the bullheads and 200 g for the pike) than those caught in September (70 g for the bullheads and 400 g for the pike), although there was a large variation in size at all sampling times. Water was extracted from the samples by freeze-drying and analyzed by LSC. The solid matter was washed with tritium-free water, oven-dried and combusted in a combustion bomb for subsequent OBT analysis by LSC.

Table 2 gives some information, taken from the literature, on nutrient composition and water equivalent factors for the fish and clams. Nutrient data could not be found for bullheads so values are given for carp, which are believed to be a reasonable surrogate. The data are for the edible portion of the organisms and may not reflect the composition of the internal organs. Table 3 shows measured water contents of the fish and clams. The total weight and organ weights of some of the fish caught in the sampling campaign of September 28 - October 1 are given in Table 4.

Table 2. Number of grams of nutrient in 100 g of edible portion of pike, carp (a surrogate for bullheads) and clams.

Nutrient	Pike	Carp (Bullhead)	Clam
Protein	18.2	18.9	10.5
Fat	1.2	7.1	1.3
Carbohydrate	0	0	3.1
Water equivalent factor	0.645	0.709	0.577

Table 3. Water contents of fish and clams.

Organism	Water content (% by weight)
Clam	89.0
Bullhead - flesh	82.3
- head	76.8
- internal organs	82.3
Pike - flesh	77.7
- head	73.9
- internal organs	81.0

Table 4. Total weight and organ weights (g) of fish caught September 28 – October 1.

Fish	Total weight	Organ				Total organ	Comments
		Liver	Gonads	Stomach	Intestines		
Pike	258	2.08	2.43	4.32	4.02	12.85	Male, stomach empty
Pike	453	4.81	5.09	6.23	7.90	24.03	Male
Pike	178	1.84	0.81	3.70	4.10	10.45	Tail damage
Pike	558	5.54	10.65	10.19	7.61	33.99	Female
Bullhead	69.1	0.50	0.22	0.66	1.02	2.400	Female
Bullhead	120	3.76	0.39	11.21	7.39	22.75	Exceptionally large male; stomach contained a sunfish

Air: The tritium in the air above Perch Lake comes primarily from evapotranspiration from the lake and the adjacent wetland. Fluxes from the wetland to the air during daytime in the summer are about 1-3 Bq m^{-2} s^{-1}. Monthly-averaged air samples were collected with passive diffusion samplers in the months of August and September at sites S1, S2 and S3. The samplers were located 1-2 m from the shoreline at a height of 1 m. Analysis by LSC provided concentrations in Bq m^{-3} air, which were converted to Bq L^{-1} air moisture using the measured average monthly temperature and an estimated relative humidity of 75%.

Uncertainties: Counting errors in the HTO concentrations in lake water, plants and aquatic animals were generally less than 2% but reached 10% in some cases of low concentrations. Total uncertainties in the HTO concentrations in sediment water were somewhat larger because of the difficulties in keeping lake water out of the sample. Replicate sediment samples from the same location showed differences of about 30%. A similar variation among individual plant and animal samples would be expected because of natural variability but may not be evident in the composite samples that were analysed. Uncertainties in air concentrations arose due to counting errors, and to uncertainties in the performance of the passive samplers and in determining the volume of air sampled. The total uncertainties in the air concentrations are estimated to be less than about 30%.

Counting errors for OBT concentrations were usually less than 5% but additional uncertainty arose due to difficulties in removing exchangeable OBT from the samples and in the combustion process. The total uncertainty in the OBT measurements is estimated to be about 20%. Differences among replicate samples from the same location may be larger because of natural variability.

INPUT DATA

Measured HTO concentrations in water, sediment water and air moisture are shown in Table 5. Where more than one value is listed for a given parameter, separate samples were taken close to the same location. Concentrations of the water and sediment samples collected from shore may not reflect concentrations in the main body of the lake. At sampling sites S1, S3 and S4, the near-shore samples were taken close to the inlets of the associated streams and concentrations may have been diluted by the relatively clean inflow. In contrast, the near-shore samples taken at S2 may be higher than those further out in the lake since concentrations in Inlet 2 are relatively high. Air concentrations were highest near S2, which is directly over the underground plume, and decreased from August to September.

Table 5. Measured HTO concentrations in water, sediment water and air moisture.

Month	Compartment	HTO Concentrations ($Bq\ L^{-1}$)				
		S1	S2	S3	S4	Outlet
May	Surface water - offshore	4350	5450	4730		
	Sediment water - offshore	4730 3330 3830	10890 13570 13210	1320		
July	Surface water - offshore - from shore near inlet	4640 4150	4590 3330	4620 3800	 91	4660
	Deep water - offshore* - from shore near inlet‡	4480 3900	4460 2570	4420 3580		4620
	Sediment water - from shore near inlet	2300	7120	70		
Sept	Surface water - from shore near inlet	2030	9290	139		
	Deep water - from shore near inlet‡	2080	9190	113		
	Sediment water - from shore near inlet	1500 1650	7420 4550	84		
	Air - August - September	740 660	1970 1770	510 260		
Nov	Surface water - offshore	3840	5270	3770		
	Deep water – offshore*	3480	9350	3770		

* collected at a depth of about 1.5 m
‡ collected at a depth of about 0.4 m

Based on single measurements made from shore at the outlet of the lake in 2001, HTO concentrations in surface water were 6330 Bq L^{-1} in June and 6660 Bq L^{-1} in December.

The rate and distribution of HTO releases to the lake are too poorly known to allow concentrations in lake water to be predicted using a model. The concentrations needed for the scenario calculations must therefore be estimated from the data in Table 5. Similarly, no information is available on rates of eutrophication, biomass production or decay in the lake, or on OBT concentrations in soils of the watershed, to help in estimating OBT concentrations in sediments.

SCENARIO CALCULATIONS

Using the information provided above, calculate

(i) HTO and non-exchangeable OBT concentrations in cattails, and in bladderwort and hornwort combined, for the May sampling period for the near-shore portions of sites S1, S2 and S3. For cattails, give concentrations for both the above water and below water parts of the plant. Also, calculate HTO and non-exchangeable OBT concentrations in algae for the May sampling period for the offshore portions of sites S1, S2 and S3. For HTO, give the results in Bq L^{-1}; for OBT, give the concentration in the combustion water (i.e., Bq L^{-1} water equivalent).

(ii) HTO and non-exchangeable OBT concentrations in clams, bullheads and pike for each of the three sampling periods. For bullheads and pike, give concentrations in head, flesh and internal organs (liver, gonads, stomach and intestines). Give the results in Bq L^{-1} for HTO and Bq L^{-1} water equivalent for OBT.

(iii) non-exchangeable OBT concentrations in sediments for the May sampling time for the near-shore portions of sites S1, S2 and S3, in units of Bq L^{-1} water equivalent.

(iv) 95% confidence intervals on all predictions in (i) - (iii).

Appendix B: Model descriptions

VNIIEF Model

Key Model Assumptions:

- The turnover time for tritiated water in the tissues of aquatic organisms is very rapid.
- Clams, pike and bullheads take in tritium through exchange with water and also through ingestion.
- All water layers are taken into account in calculating concentrations in pike, which migrate throughout the lake.
- Bullhead is a bottom fish so that only deep water is taken into account in calculating concentrations.
- Clams live at the lake bottom but take up detritus formed in all water layers.

The parameter values used in the calculations and further assumptions are based on the work of Murphy (1993) and Diabate and Strack (1993).

Model Description:

The table below provides a short description of the model. k is the isotopic discrimination factor in OBT formation.

<table>
<tr><th colspan="2" rowspan="2">Compartment</th><th colspan="2">Tritium source</th><th rowspan="2">Comment</th></tr>
<tr><th>HTO</th><th>OBT</th></tr>
<tr><td colspan="2">Bladderwort and hornwort</td><td>Surface water</td><td>Surface water (k = 0.8)</td><td></td></tr>
<tr><td rowspan="2">Cattails</td><td>Above water</td><td>Air and sediment water</td><td>Sediment and surface water (k = 0.8)</td><td>HTO concentration is calculated from $C_{HTO} = 0.75C_{air} + 0.25C_{sediment}$; $C_{air} = 0.9C_{surf}$</td></tr>
<tr><td>Below water</td><td>Sediment water</td><td>Sediment water (k = 0.8)</td><td></td></tr>
<tr><td colspan="2">Algae</td><td>Lake water</td><td>Lake water (k = 0.5-0.8)</td><td>Algae are assumed to access all water layers, which have an effective concentration of $C = 0.5(C_{surf} + C_{deep})$</td></tr>
<tr><td rowspan="3">Pike</td><td>Head</td><td>Essentially lake water</td><td>Lake water (k = 0.1-0.5)</td><td rowspan="3">Concentration in lake water is calculated as $C = \frac{1}{N}\sum_i 0.5(C_{surf} + C_{deep})$, where the summation is over sample points.
OBT concentration in the diet is equal to the concentration in lake water.</td></tr>
<tr><td>Flesh</td><td>Lake water and water in diet</td><td>Lake water (k = 0.1-0.5) and diet (k = 0.5).</td></tr>
<tr><td>Internal organs</td><td>Water in diet</td><td>Diet (k = 0.5)</td></tr>
<tr><td rowspan="3">Bullhead</td><td>Head</td><td>Essentially lake water</td><td>Lake water (k = 0.1-0.5)</td><td rowspan="3">Concentration in lake water is calculated as $C = \frac{1}{N}\sum_i C_{deep}$
OBT concentration in the diet is equal to concentration in lake water.</td></tr>
<tr><td>Flesh</td><td>Lake water and water in diet</td><td>Lake water (k = 0.1-0.5) and diet (k = 0.5).</td></tr>
<tr><td>Internal organs</td><td>Essentially water in diet</td><td>Diet (k = 0.5)</td></tr>
</table>

Clams	Sediment and deep water	Deep water (k = 0.1- 0.5) and diet (k = 0.5)	Concentration in lake water and diet are calculated as $C_{water} = \frac{1}{N}\sum_i C_{sediment}$, $C_{diet} = \frac{1}{N}\sum_i \frac{1}{3}\left(C_{surf} + C_{deep} + C_{sediment}\right)$
Sediment	-	Surface water in May	The detritus that makes up sediments is considered to form in May in surface water.

References

Murphy, C.E. 1993. Tritium transport and cycling in the environment. Health Physics 65, 683-697.

Diabate, S. and S.Strack. Organically bound tritium. Health Physics 65, 698-712.

EDF Model

The EDF model for aquatic contamination is a dynamic model that computes concentrations of HTO and OBT in phytoplankton and fish. Although freshwater macrophytes, molluscs and sediment are not included in the EDF model, concentrations were calculated for all the compartments covered in the Perch Lake scenario.

(i) Concentrations in aquatic plants

The general assumption is that HTO in plants is equal to HTO in the surrounding environment.

For algae, the water concentration to consider is the offshore surface concentration. OBT concentrations were calculated on the assumption that the OBT/HTO ratio is equal to 0.6 in aquatic plants (Kirchmann et al. 1979).

For worts, the water concentration to consider is the near-shore surface concentration. But these data are not available. The July measurements indicate that the ratio "near-shore surface water/offshore surface water" was equal to 0.8. Using this ratio, the near-shore surface water in May was estimated from the offshore surface water in May. OBT was calculated on the assumption that the OBT/HTO ratio equals 0.6 in aquatic plants.

For cattails, the HTO concentration in the below-water parts was assumed to be identical to the concentration in worts. In contrast, the OBT concentration in the below-water parts was assumed to be in isotopic equilibrium with shore sediment water and surface water. The shore sediment water concentrations for a given site were estimated from the measurements made at that site in July and October. HTO concentration in the above-water parts of cattails was assumed to equal the average of the HTO concentrations in air and shore surface water, the latter being calculated in the same way as for worts. Half of the OBT in the above-water parts was assumed to come from the below-water parts with the other half being in isotopic equilibrium with HTO in the above-water parts. None of the OBT calculations for cattails took into account the OBT/HTO ratio of 0.6.

(ii) Concentrations in animals

HTO in clams was considered to be in isotopic equilibrium with HTO in sediment water, calculated as the average of the near-shore and offshore sediment concentrations at the three sampling sites. The OBT/HTO ratio was set to 0.45 (Kirchmann et al. 1979).

HTO in both bullheads and pike was assumed to be in isotopic equilibrium with the offshore surface water, calculated as the average over the three sampling sites. OBT in fish was assumed to be controlled by the tritium transfer rate between food and fish and by the specific food intake rate (10^{-2} day^{-1}). Moreover, the concentration in food was a function of the concentration in water. Thus, the EDF model is based on a transfer rate from water to fish that is consistent with an apparent OBT/HTO ratio of 0.45 (Kirchmann et al. 1979). To account for the size difference between bullheads and pike, pike were assumed to be older and to have grown in water with an HTO concentration of 6000 Bq/L in previous years.

(i) Concentrations in sediment

OBT in sediment has a very slow turnover rate and is a function of OBT in aquatic biota and OBT in terrestrial organic matter that finds its way into the lake. Since concentrations from previous years either in the terrestrial or aquatic environment were not available, the following assumptions were made:

- The spatial distribution of sediment OBT is similar to the spatial distribution of lake HTO. Thus concentrations at S2 exceed those at S1, which exceed those at S3.
- OBT concentrations in aquatic biota in previous years could be as high as the upper limit of the 95% confidence interval for water concentration in 2002 (see section 'iv'). This value was assigned to the sediment OBT concentration at S2.
- A fraction of the sediment OBT comes from lake plants and animals growing in 2002. This value was assigned to S1.
- The lowest OBT concentration, the value assigned to S3, was derived by assuming that the sediment is in isotopic equilibrium with organic matter resulting from the decomposition of terrestrial vegetation that finds its way into the lake. The concentration in terrestrial vegetation was calculated from the air concentration near S3 and an OBT/HTO ratio of 0.6.

Hence, the differences in sediment OBT concentrations among sampling sites do not depict actual spatial variations but represent different origins of sediment OBT more or less randomly assigned to each sampling site.

(iv) 95% confidence intervals

The largest source of uncertainty was the heterogeneity in water concentrations. The 95% confidence interval of a lognormal distribution fitted to the observed water concentrations had upper and lower limits of 1850 and 8745 Bq/L. This interval was used as the uncertainty in the HTO concentrations in all plants and animals with the exception of the above-water parts of cattails, for which the lower limit became the observed atmospheric HTO concentration.

For OBT, a second source of uncertainty is the OBT/HTO ratio, which could vary between 0.4 and 0.9 for plants and between 0.1 and 0.9 for animals. The lower limit of the OBT confidence interval was found by multiplying the lower limit of the confidence interval for water concentrations by the lower value of the OBT/HTO ratio. A similar procedure was used to estimate the upper limit. This is not, in its strictest sense, a 95% confidence interval.

For sediment, the uncertainty range represents the absolute maximum and minimum values corresponding to the different OBT origins: OBT from 'old organic matter' in S2, OBT from aquatic biota growing in 2002 in S1, and OBT from terrestrial organic matter in S3.

References:

Ciffroy, P., F. Siclet, C. Damois and N. Luck. 2006. A dynamic model to assess radiological consequences of tritium routinely released in rivers. Submitted to Journal of Environmental Radioactivity.

Kirchmann, R., S. Bonotto, S.D. Soman, T.M. Krishnamoorthy, T.S. Iyengar and A.A. Moghissi. 1979. Transfer and incorporation of tritium in aquatic organisms. In "Behaviour of Tritium in the Environment", IAEA-SM-232/96.

SRA Model

The calculations for the Perch Lake scenario are based on the assumptions that the tritium flux into the lake is stationary and that the free-water tritium (FWT) and OBT in the plants and animals are in equilibrium with an effective HTO concentration C_{eff}, defined as the average HTO concentration in the lake water or the atmospheric water vapor to which a fish or plant is exposed over its growing period. C_{eff} depends on location and time and, for fish, on the time spent under given living circumstances. At equilibrium, the FWT concentration in a living organism (C_{FW}) is equal to C_{eff}:

$$C_{FW} = C_{eff} = \sum_i F_i \times C_i$$

where F_i = fractional contribution to the effective tritium concentration from the *i*th HTO source, and
C_i = tritium concentration at equilibrium in the *i*th HTO source.

The OBT concentration is given by

$$C_{OB} = FC \times C_{FW}$$

where FC is a discrimination factor for tritium in organic material. In the absence of experimental data for the organisms considered in the Perch Lake scenario, published values of FC for other animals or plants grown in aquaria or pools were used. The values of F_i and FC used in the calculations are shown in Table 1.

The determining factors for the FWT and OBT levels in algae and worts were assumed to be the near-shore lake water tritium concentration at the sampling time and the tritium discrimination factor in photosynthesis. The HTO concentration in below-water cattails was assumed to be controlled by the near-shore sediment concentrations. For the above-water parts of cattails, the tritium concentration in atmospheric water vapor also played a role. The fractional contribution of the atmospheric HTO was assumed to be 0.3 but was neglected in the present calculation. Thus, the FWT concentration in the above-water cattails was set equal to 0.7 times the near-shore sediment concentration. Since information on the near-shore sediment water concentration was not available for May, the data for July were used instead.

For each animal species, an effective lake water concentration was estimated from the average tritium concentration in the different parts of the lake weighted by the time the animals spent in each part. For bullheads, lake water and sediment water were assumed to contribute equally to the body FWT, since the fish spends its time near the sediment/water interface. For pike and clams, the lake-averaged HTO concentration was taken as the effective HTO concentration. The tritium discrimination factor FC for OBT was obtained in analogy with published values for other species.

Sediment OBT concentrations at Site S3 were assumed to be equal to 0.63 times the HTO concentration in the near-shore sediment water.

It was rather difficult to determine the confidence level for the model predictions since there are so many sources of uncertainty. The uncertainty estimates reflect solely the standard deviation of the effective tritium concentration estimated for individual samples, based on the observed variation of tritium levels in the lake or sediment water.

Table 1. Factors determining tritium concentrations in the endpoints of the Perch Lake scenario

Endpoint	Tritium source	Fractional contribution to effective tritium concentration (F_i)	Averaging time	OBT specific activity relative to effective tritium concentration (FC)
Algae	Near-shore surface water	1.0	Sampling period	0.7
Wort	Near-shore surface water	1.0	Sampling period	0.7
Cattail above water	Near-shore sediment water Atmosphere	0.7 0.3	Sampling period	0.5
Cattail below water	Near-shore sediment water	1.0	Sampling period	0.5
Clam	Lake averaged*	1.0	Sampling period	0.5
Bullhead	Lake averaged* Offshore sediment water	0.5 0.5	Sampling period	0.64 (head), 0.70 (flesh), 0.66(organs)
Pike	Lake averaged*	1.0	Sampling period	0.64 (head), 0.70 (flesh), 0.66(organs)
Sediment	Near-shore sediment water	1.0	Year	0.63

* The lake-average HTO concentration was assumed to be given by the average HTO concentration of the deep and surface waters at Sites S1 and S3 and the sediment water at Site S3.

BioM Model

The aim of the BioM model is to improve the estimation of long-term tritium doses by re-evaluating the way in which OBT is treated. The model calculates the concentration of buried tritium rather than the tritium traditionally considered to be organically bound. Buried tritium is tritium in exchangeable positions in large molecules that becomes hidden from the effects of washing when the free water of the sample is extracted by freeze-drying or azeotropic distillation. This tritium appears as part of the experimental yield when the sample undergoes a traditional analysis for OBT, but is converted to HTO as soon as it is ingested and so does not contribute to the OBT dose. Improved understanding of the amount of buried tritium that forms in plant and animal species will lead to improved dose estimates from OBT.

HTO Concentrations: The HTO concentration in each scenario endpoint was assumed to equal the HTO concentration in the air, water or sediment compartment to which the plant or animal was exposed (Table 1). Bullheads and pike were assumed to move everywhere in the lake.

Table 1. Compartments to which a given endpoint is exposed

Endpoint	Compartment
Algae	Local offshore surface water
Worts	Arithmetic mean of local offshore sediment and surface water
Submerged cattails	Local sediment water
Emergent cattails	Local sediment water multiplied by 1.1 to account for the difference in vapour pressure between HTO and water vapour
Clams	Arithmetic mean of S1 and S2 offshore sediment water
Bullheads and pike	Offshore surface and sediment water averaged over the 3 sampling sites

OBT Concentrations: The experimental basis of the BioM model is the observation that freeze-drying or azeotropic distillation of a sample to extract the free water results in a large part of the exchangeable tritium becoming non-exchangeable in OBT analysis (Baumgärtner and Donhaerl 2004). The tritium is “buried” inside the biopolymers or in shell water that is separated from bulk water (Falk et al. 1970). Shell water does not freeze at temperatures of dry ice or liquid nitrogen. Accordingly, the BioM model assumes that OBT measured using traditional methods consists of three components:

$$C_{OBT} = C_{CBT} + C_{OBTex} + C_{SBT}, \qquad (1)$$

where C_{CBT} is carbon bound tritium, C_{OBTex} is tritium that is nominally exchangeable but buried by freeze drying or azeotropic distillation, and C_{SBT} is tritium buried in water molecules of the solvation shells. C_{CBT} is formed by photosynthetic and enzymatic pathways and is the quantity that determines the long-term radiation dose from tritium. According to Eq. (1), $C_{CBT} \sim (C_{OBT} –$

C_{OBTex}), and C_{CBT} can be determined from analytical measurements of C_{OBT} if C_{OBTex} can be estimated. The BioM model provides a way to calculate C_{OBTex}.

The starting point of the calculation is the HTO concentration in the tissues (C_{HTO}, Bq/L) and the proportion of carbohydrates, proteins and DNA in the tissues (Baumgärtner 2005). Then the concentration of buried tritium (C_{OBTex}, Bq/L) is given by

$$C_{OBTex} = \alpha\, C_{HTO}\, (18/2)\, [\sum_i C_H H_{ex}]/W_{eq} \qquad (2)$$

where α is the T/H fractionation factor of tritium between water and exchangeable hydrogen positions,
C_H is the hydrogen content (fraction),
H_{ex} is the fraction of exchangeable hydrogens, and
W_{eq} is the water equivalent.

The summation in Eq. (2) is over carbohydrates, proteins and nucleotides. The product C_H x H_{ex} has a value of 0.019, 0.017 and 0.0057 for carbohydrates (Di Bari et al. 2003), proteins (Klapper 1977) and nucleotides (Baumgärtner 2005), respectively. The model does not take into account tritium that accumulates in the hydration shells, which remains with the organic matter following freeze drying, so that the predictions may underestimate C_{OBTex} concentrations by 20 to 40%.

The value of the tritium fractionation factor is uncertain. $\alpha \sim 1.4$ is valid for 1-step exchange reactions and $\alpha \sim 1.4^2 \sim 2$ for 2-step reactions. DNA shows both values. $\alpha \sim 1.4$ is found in the first DNA-hydration shell and $\alpha \sim 2$ in the base pairing H-positions inside DNA (Baumgärtner and Kim, 2000). Since the dominant type of H/T exchange reaction for aquatic systems is unknown, both values were used in the calculations. For simplicity, all plants were assumed to be made up of carbohydrates only and all animals of proteins only.

The 95% confidence intervals were calculated with 1 degree of freedom by the t-distribution assuming 2% standard deviation of the mean (7% in the case of clams and cattails because they are supplied with HTO from the sediments).

C_{OBTex} makes up a substantial proportion of C_{OBT}. Furthermore, the amount of tritium unaccounted for in the solvation shells (0.25 to 0.75g $g_{H2O}/g_{protein}$ (Saenger 1987) and up to 0.3g_{H2O}/g_{starch} (Di Bari et al. 2003)) and the large primary kinetic isotope effects in enzyme-catalyzed reactions (Cleland and O'Leary 1977) suggest strongly that $C_{CBT} < 0.1\, C_{OBT}$ if freeze drying or azeotropic distillation is used to extract the free water from the sample prior to analysing for OBT.

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IFIN Model

Plants

Based on experimental evidence, HTO concentrations in plants are in equilibrium with local HTO concentrations in water. For the emergent parts of cattails, the role of transpiration and exchange with atmospheric water is also considered. An average HTO concentration for each plant type was determined by averaging over the water column and over the three sampling sites. The concentrations in bottom water given in the scenario description were supplemented with data from Cornett et al. (1989). For worts and below-water cattails, the data used in the average were the near-shore concentrations measured in July. These were averaged with the August air concentrations to give an HTO concentration in above-water cattails. The concentration in algae was estimated from the average water concentration at the site where the algae were collected.

OBT in aquatic plants is produced as in terrestrial plants but at a slower rate, which implies that OBT concentrations should be based on average HTO levels in water for the month or so before sampling. In the absence of this information, the OBT concentrations in worts and cattails were found by multiplying the plant HTO concentrations for May by 0.8. OBT concentrations in algae ($C_{o,phpl}$, Bq/kg fw) were calculated using a model for tritium dynamics in the aquatic environment (Galeriu et al. 2005):

$$\frac{dC_{o,phpl}}{dt} = 0.4 \cdot \mu \cdot Dryf \cdot C_W - \mu \cdot C_{o,phpl} \quad (1)$$

where μ and Dryf are respectively the growth rate (per day) and dry mass fraction of the algae and C_W is the HTO concentration in water (Bq/L). The growth rate is given by (Ray, 2001)

$$\mu = 0.75 * (3 - 0.3 * \log(V_p)) \quad (2)$$

where V_p is the cell volume, which can range from 10 and 10^7 μm^3. Assuming the algae belong to a typical class of the phylum Chlorophyta, Eq. (2) gives a growth rate of 1.8 d^{-1} in full light. A growth rate near 0.5 d^{-1} is assessed for the conditions of the scenario. With this value, and an assumed water equivalent factor of 0.6, the OBT concentration in the algae was found as the steady-state solution to Eq. (1).

Animals

Based on experimental evidence, HTO concentrations in animals are in equilibrium with local HTO concentrations in water. A nominal value for the water concentrations to which clams and bullheads were exposed was deduced from an overall assessment of the bottom water and sediment concentrations over time throughout the lake. The HTO concentration in clams and bullheads was assumed equal to this water concentration, with some seasonal variation introduced in the case of bullheads. Similarly, HTO concentrations in pike were set equal to the water concentrations averaged over the water column and over the three sampling sites for each sampling time.

OBT concentrations in aquatic producers (C_{OBT}, Bq/kg fresh weight) are given by

$$\frac{dC_{OBT}}{dt} = a\,K_1 C_f + b\,K_w C_w(t) - K_{0.5} C_{OBT} \qquad (3)$$

where a is the assimilation factor for OBT from food,
b is the water to OBT transfer factor,
K_1 is the food uptake rate (kg kg^{-1} d^{-1}),
K_w is the water uptake rate (m^3 kg^{-1} d^{-1}),
$K_{0.5}$ is the OBT elimination rate (d^{-1}),
C_f is the OBT concentration in food of zooplankton (Bq/kg fresh weight), and
C_w is the HTO concentration in water (Bq/m^3).

The constants a and b in Eq. (3) were established using measured specific activity ratios of OBT in the organism of interest and OBT in food or HTO in water (Table 1). Elimination rates were assessed from experimental metabolic data.

Table 1. Specific Activity Ratios (SAR) for different organisms

	SAR (HTO source)	SAR (OBT source)
Zooplankton	0.4	0.6
Molluscs	0.2	0.8
Crustaceans	0.2	0.8
Planktivorous fish	0.2	0.8
Piscivorous fish	0.2	0.8
Terrestrial mammals	0.25	0.75

Clams are filter feeders, eating phytoplankton and zooplankton but also retaining detritus. OBT in clams is due to OBT in the food they eat but also due to conversion of HTO. Both types of plankton have low OBT halftimes (less than 6 days) so OBT in the food will closely follow the dynamics of HTO in water, but with less variability. The OBT loss rate of clams is in the range of 40-100 days and will reduce the dynamics of OBT in clams. The uncertainty in the predicted OBT concentrations in clams is large because critical information on OBT concentrations is missing.

Bullheads are benthic fish eating mostly zoobenthos, zooplankton, invertebrates and detritus, as well as fish and plants. The exact diet depends on the age of the fish and their environmental conditions. They are abundant in areas with submerged plants. Bullheads have a variable metabolic rate, especially near a mass of 100 g. For the May harvest, when the bullheads had an average mass of 40 g, the OBT half time was estimated to be 20-40 days. In September, when the average mass was 70 g, this increased to 25-50 days. Estimates of OBT concentrations in bullheads are difficult to make because the information needed to assess OBT in sediments is missing. OBT concentrations in viscera may be slightly higher than in flesh but some of the key data needed to make a quantitative assessment were not available.

Pike are pelagic fish that eat other fish. OBT concentrations in viscera may be higher than in flesh.

Sediments: A series of papers by Hakanson and Bullion (2002) on biomass, turnover times and biomass loss rates in freshwater systems suggests that most OBT in sediments comes from benthic algae and macrophytes, and is sensitive to concentrations in bottom water. This information was used to develop nominal estimates of OBT concentrations in sediments.

Uncertainty: The key information required to estimate the various endpoints is the HTO water concentration for each sampling time, site and organism. The scenario does not offer enough detail of this kind and this is the main source of uncertainty. An additional difficulty in assessing the confidence interval on OBT concentrations is the fact that the rate at which tritium enters the lake as OBT is not known. The estimated confidence intervals are based on judgment.

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Japanet Model

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<u>Assumptions:</u>

1. No special numerical models or transfer parameters for tritium uptake by plants or animals were used in the calculations.
2. The HTO concentration in lake water is not homogeneous and varies with location and season. The concentrations in plants and animals will also vary with time and space.
3. The tissue free water tritium (TFWT) in plants and clams is equal to the HTO concentration in lake water taken at the time and place the samples were collected.
4. The TFWT of fish is equal to the HTO in lake water averaged over the whole lake in all seasons.
5. The non-exchangeable OBT concentration (nOBT) of every plant and animal is 80% of its TFWT concentration.
6. There is no difference in the nOBT concentration in different parts of the fish.
7. The nOBT of the sediments is the mean value of the nOBT of plants and animals.
8. The “95% confidence interval” for a given endpoint is assumed to be ±20% of the HTO concentration in lake water used to predict that endpoint.
9. Perch Lake is a well-mixed aquatic environment in which the nOBT of living species has reached steady-state.

GE Healthcare Model

Basic assumptions

The model is based on 1 kg of fish, as this quantity can be easily amplified to the amount needed for consumption of fish within the critical group. The fish has been divided into two compartments, a fish fast compartment and a fish slow compartment (Figure 1). The fish fast compartment represents the tissue free water inside the fish, whilst the fish slow compartment represents the organic matter of the cells. It is assumed that these two compartments are in equilibrium within the fish. Transfer from the fish fast compartment to the fish slow compartment is anabolism, a constructive metabolic process that synthesizes more complex molecules. Transfer from the fish slow compartment to the fish fast compartment is catabolism, degredative chemical reactions in cells that convert polymer metabolites via monomers. The model is time-dependent and was developed for a marine environment, and had to be modified to treat the Perch Lake scenario, which involves a freshwater ecosystem. Since the model is geared to predicting concentrations in fish rather than aquatic plants, no calculations were carried out for worts or cattails.

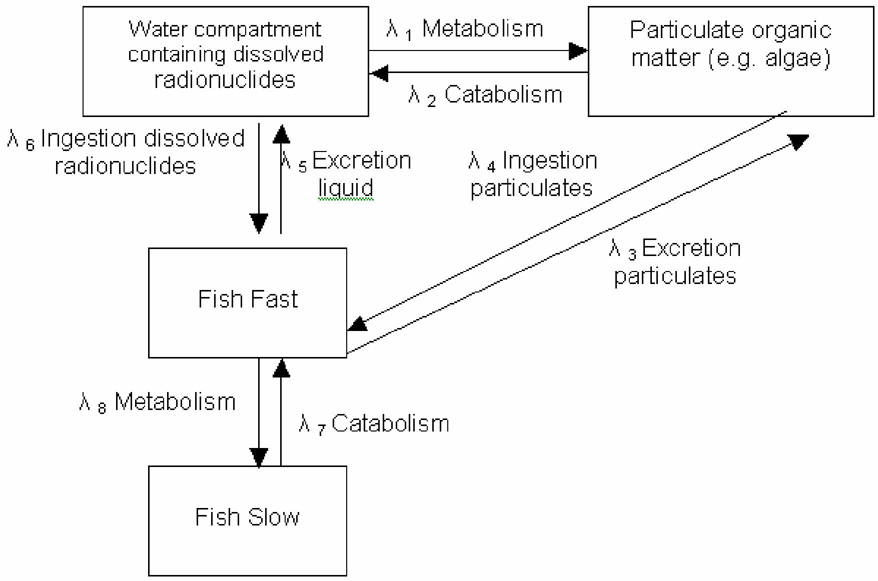

Figure 1 Dynamic Fish Conceptual Model

Concentration of tritium in water: There was no information in the scenario description on the flux of tritium into or out of the lake, which is the starting point of the GE model. Therefore the model was run with nominal concentrations in one inlet and one outlet and the results were tuned so that modeled concentration in the water column matched the measured concentration as given in the scenario.

Algae: No information was given in the scenario on algal growth rate or turnover rate. Algae were therefore taken to be broadly similar to bacterial particulate organic matter in the GE model, as some green algae are unicellular and reproduce asexually by fission (splitting) or fragmentation. Thus algal growth rate and transfer between the algae and the water column was based on rates of metabolism and catabolism for particulate matter in the GE model, which, in turn, is based on bacterial physiology.

Fish and Clams

Clams were taken to be similar to mussels in the GE model, ingesting water and algae (particulate organic matter). Bullheads and pike were taken to be similar to flounder in the GE model.

Ingestion: It is assumed that the hydrogen and carbon content in 1kg of fish is the same as the hydrogen and carbon content in the material that 1kg of fish ingests. It has been found that 1kg of fish is made up of 30% carbon, 47% hydrogen and 23% oxygen. Fish (and mussels) were taken to consume 1% of their body weight per day (Craig and Helfrich 2002). Therefore it was assumed that 1 kg of fish consume 3.65 kg H per year. The ratio of flux/inventory of the donor compartment for tritium was assumed to be the same as for hydrogen.

Tritium has two other routes of ingestion into the fish, as tritiated water from the water compartment that the fish inhabits and as particulate OBT associated with the suspended material in the water compartment. Of the tritium ingested, 20% is ingested via inspiration (as dissolved tritium) and 80% via diet (as particulate tritium) (Rogers 1996; McCubin and Leonard 2001).

Excretion: All the tritium taken up into the fish is released back into the water from the fish fast compartment via excretion, expiration and death. Excretion is divided into what is excreted in particulate form as faeces and that excreted via expiration. It was assumed that of the hydrogen excreted, 90% is in dissolved form and 10% is particulate matter (Arapis 1987; CEFAS Report).

Metabolism rate: Using a cautious approach, it is assumed that 3% of the intake of tritium into the fast compartment (from both the water compartment and particulate suspended material compartment) is incorporated into the organic constituents of the fish in the slow compartment due to growth (Hamby and Palmer 2001; Craig and Helfrich 2003).

Catabolism rate: A constant net transfer rate was assumed; therefore there were no transfer losses from one compartment to the next. All that enters the fast compartment is lost at the same rate because the fish is in dynamic equilibrium.

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LIETDOS_W MODEL

The LIETDOS_W model was developed to predict levels of radioactivity in water sediment, fish and aquatic plants in lake ecosystems. LIETDOS_W is a dynamic linear compartment model that is described by first order differential equations with constant or time-dependent coefficients. This model has been developed at the Institute of Physics (Lithuania) and used to evaluate the contamination in the Ignalina NPP cooling pond (Druksiai Lake). In the case of the Perch Lake scenario, an additional submodel was developed for predicting non-exchangeable OBT concentrations. The parameters associated with this new submodel are presented in Table 1.

Table 1. Submodel parameter values

Endpoint	$HTO_{endpoint}$ / HTO_{water}		OBT / HTO		References
	Mean value	Range	Mean value	Range	
Worts	0.9	0.8-1.0	0.75	0.4-1.0	1, 2
Cattail above water	0.75	0.6-0.9	0.66	-	1, 2
Cattail below water	0.9	0.8-1.0	0.82	0.5-1.0	1, 2
Clam	0.9	0.8-1.0	0.38	0.15-0.6	1, 2
Bullhead	0.9	0.8-1.0	0.38	0.15-0.6	1, 2
Pike	0.9	0.8-1.0	0.38	0.15-0.6	1, 2

The prediction of HTO and OBT concentrations in cattails, bladderworts and hornworts was based on measured HTO offshore surface water concentrations at sampling sites S1, S2 and S3 in May. HTO concentrations in all aquatic plants and animals were assumed to be equal to or slightly less than the corresponding water concentration [1]. The OBT/HTO ratio varied between 0.66 and 0.82 for worts and cattails and was 0.38 in the case of fish and clams [1, 2]. The modeling of algae and sediments was beyond the capability of the model.

The standard deviation of HTO concentration in lake water was calculated according the data given in the scenario description. In the case of other endpoints, the standard deviation was evaluated according to the data presented in Table 1. The 95% confidence intervals were calculated using lognormal distributions by means of Crystal Ball software.

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