EMRAS Tritium/C14 Working Group

THE PICKERING SCENARIO

Final Report
April 2008

1. SCENARIO DESCRIPTION

This scenario is based on data collected in the vicinity of Pickering Nuclear Generating Station (PNGS), a collection of eight CANDU reactors on the north shore of Lake Ontario. The surrounding environment contains slightly elevated levels of tritium due to continuous, routine discharge from the reactors. The releases have been going on for many years and concentrations in various parts of the local environment are likely to be in equilibrium. A large number of environmental and biological samples were collected in 2002 from four sites in the vicinity of the station. HTO concentrations were measured in air, precipitation, soil, drinking water, plants (including the crops that make up the diet of the local farm animals) and products derived from the animals themselves; OBT concentrations were measured in the plant and animal samples. These data were used as a test of models that predict the long-term average tritium concentrations in terrestrial systems due to chronic releases.

The samples were taken at two dairy farms (DF8 and DF11), a hobby farm (F27) and a small garden plot (P2) (Figure 1). All of the sampling sites were located to the northeast of PNGS; the two dairy farms lay about 10 km from the station, the hobby farm about 7 km and the garden plot about 1 km. The two dairy farms yielded much the same sort of samples, including pasture grasses, a variety of grains, milk and meat. In contrast, F27 produced mainly fruit, garden vegetables, chickens and eggs. A limited number of plants are grown at P2 for research purposes and raspberry leaves and grass were sampled.

The cows at DF8 and DF11 were fed total mixed ration (TMR), a blend of various feeds harvested in the previous year. Most components of the mixture were obtained locally. Estimates of the total food intake by the cows were available from the owners. The chickens raised at F27 were essentially free-range and their food intake was not regulated or monitored. As a result, the make-up of their diet and their intakes could only be estimated. The amount of drinking water ingested by the cows and chickens was not monitored.

Tritium concentrations in air and precipitation were available from a monitoring program carried out by the utility. Air concentrations at P2 were measured monthly using an active air sampler, and were considered reliable. However, at DF8, DF11 and F27, air concentrations were available only as annual averages from passive diffusion samplers. For a number of reasons, these data were considered untrustworthy and were replaced with the predictions of a sector-averaged atmospheric dispersion model that produced concentrations in good agreement with the observations at P2, DF8 and DF11.

All of the other samples were collected in two field campaigns conducted in 2002, the first from July 8 - 10 and the second from September 16 - 18. All of the samples collected in July were dried before the HTO could be extracted and so were suitable for OBT analysis only. The September samples were frozen in their fresh state and were analysed for both HTO and OBT. At the dairy farms, samples were collected of each of the plants that made up the animal diets, as well as separate samples of TMR. At F27, additional measurements were made of garden vegetables, root crops and fruit. The meat samples from DF8 and DF11 came from calves that were either stillborn or died from complications at birth. The mothers were three years old or younger and were raised exclusively on these farms. Additionally, composite milk samples consisting of a mixture of milk from all cows in the herd were collected in July at both farms. The only animal products sampled at F27 in the July campaign were eggs. In September, in addition to eggs, blood and flesh were also analysed from a single chicken. Samples of water were taken from the deep wells that supply drinking water for the cows at farms DF8 and DF11 in the September sampling period. The concentration in drinking water at F27, which comes from a shallow well, was available as a six-month average from the routine monitoring program carried out by the utility. Soil cores were collected at a single location at each site from undisturbed grassed areas or where the soil had lain fallow for some time.

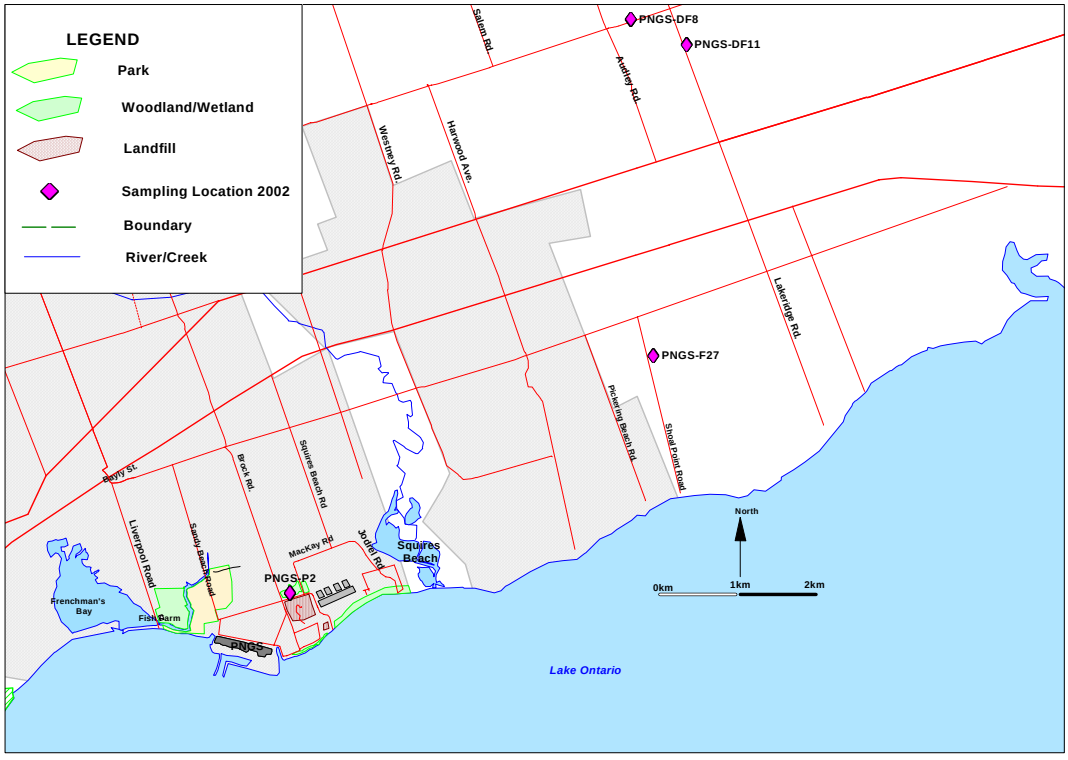

Fig 1. Map of the study area showing PNGS and the sampling sites.

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

(i) HTO and non-exchangeable OBT concentrations in the sampled plants and animal products for each site and each sampling period.

(ii) HTO concentrations in the top 5-cm soil layer for each site and each sampling period.

(iii) 95% confidence intervals on all predictions.

The full scenario description is given in Appendix B.

2. OBSERVATIONS

2.1 Measured Concentrations: Estimates of HTO concentrations in air and drinking water are shown in Table 1; HTO concentrations in monthly precipitation are given in Table 2. These are the concentrations that were supplied to the participants to drive their models. Observed concentrations in soil, plants and animal products, which were the endpoints of the scenario, are given in Tables 3, 4 and 5, respectively. The OBT concentrations are given in units of Bq L^{-1} of combustion water.

The observed concentrations in all environmental compartments were relatively low, although they were at least a factor 4-5 above background. Counting errors for both HTO and OBT samples were less than 10% in most cases. An additional uncertainty of about 30% must be added to the plant and animal concentrations to account for natural variability. A further error of perhaps 50% must also be added to the air concentrations at DF8, DF11 and F27, which were estimated using an atmospheric dispersion model.

Table 1. Measured HTO concentrations in air and drinking water. The air concentrations include a background contribution of 0.19 Bq m^{-3}

Compartment	DF8	DF11	F27	P2
Air concentration (Bq m^{-3})				
2002 May	1.01	1.01	1.56	24
June	1.39	1.39	2.14	33
July	0.93	0.93	1.43	22
August	0.88	0.88	1.36	21
September	0.67	0.67	1.04	16
Air concentration (Bq m^{-3})				
2001 May	0.49	0.49	0.77	12
June	2.83	2.83	4.40	69

July	0.86	0.86	1.34	21
August	1.23	1.23	1.92	30
September	0.66	0.66	1.02	16
Drinking water concentration (Bq L^{-1}) 2002 September	18.6	21.1	24.3*	Not relevant

* average value for June-December 2002

Table 2. Measured monthly HTO concentration in precipitation in 2002

Month	HTO Concentration in Precipitation (Bq L^{-1})		
	DF8	F27	P2
January	not available	not available	3670
February	not available	18	1350
March	not available	24	347
April	24	29	474
May	69	14	525
June	85	61	579
July	9	14	205
August	49	19	442
September	13	22	452

Table 3. Measured HTO concentration in soil water for the September sampling period

Site	Soil water concentration (Bq L^{-1})
DF8	22.5
DF11	18.7
F27	32.9
P2	552

Table 4. Measured HTO and OBT concentrations for the sampled crops

Crop type	Site	Month	Plant type	Concentration (Bq L^{-1})	
				HTO	OBT
Forage	DF8	July	Hay[¤]	-	79.4
			Haylage[¤]	-	82.0
		September	Alfalfa	21.4	25.7
			Baled hay[¤]	46.5	17.2
			Haylage	86.7	23.5
			Corn silage	31.0	25.0
	DF11	July	Alfalfa	-	43.9
			Baled hay	-	20.2
			Haylage	-	46.5
		September	Alfalfa	22.2	31.0
			Baled hay	27.8	22.2
			Haylage	10.6	31.3
			Corn silage	20.5	31.9
	F27	July	Grass	-	31.0
		September	Grass	30.2	20.3
	P2	September	Grass	2253	730
			Raspberry leaves	1564	677
Grain	DF8	July	Barley	-	50.8
		September	Feed Corn	76.0	28.5
			Barley	72.1	40.1
	DF11	July	Feed corn	-	27.9
		September	Feed corn	163.8	20.8
	F27	July	Spring wheat	-	27.4
		September	Feed corn	34.8	15.6
			Spring wheat	38.9	26.9
Total Mixed Ration	DF8	July	TMR*	-	42.5
		September	TMR	38.7	26.1
	DF11	July	TMR*	-	38.4
		September	TMR*	38.2	22.5
Root crops	F27	July	Mixed vegetables[‡]	-	42.0
	F27	September	Carrots and potatoes	38.5	40.6
			Beet	30.7	17.2
			Garlic	-	40.9
Fruit and fruit vegetables	F27	September	Tomato	35.5	27.0
			Cucumber	-	54.0
			Soya meal	61.5	20.3
			Apple	38.7	30.9
			Pear	-	38.6
			Raspberry	-	24.5

[¤] hay refers to fresh cut pasture; baled hay is dried pasture; haylage is hay that has been stored in a silo

* produced in 2001

[‡] beet, cabbage, hot pepper, onion, dill, potato, spinach

Table 5. Measured HTO and OBT concentrations in the sampled animal products

Site	Month	Animal product	Concentration (Bq L^{-1})	
			HTO	OBT
DF8	Jul	Milk	-	33.9
	Sep	Calf flesh	27.5	31.3
		Calf heart	26.9	26.9
DF11	Jul	Milk	-	21.3
	Sep	Calf flesh	29.4	32.8
		Calf heart	33.2	20.0
F27	Jul	Egg	-	44.0
		Composite egg	-	23.1
		Immature egg	-	19.1
	Sep	Egg	33.7	26.2
		Chicken blood	33.5	21.8
		Chicken flesh	-	20.3

Discussion of Observations: For the plant samples, a quantitative comparison between predictions and observations will be made for the OBT concentrations only. The HTO concentrations in plants reflect conditions in the few hours before sampling. In contrast, the air concentrations that control tritium levels in plants are available in the scenario only as averages over a month at least. This means that the predicted HTO concentrations in plants must also be averages over the growing season. This mismatch in averaging times implies that no meaningful conclusions can be drawn from a comparison of predicted and observed HTO concentrations in plants. Rather, the predictions will be used to help explain differences among model results for OBT concentrations. On the other hand, the residence time for HTO in soil and animal products is a few days and for OBT in plants and animals a few weeks, sufficiently long that concentrations in these compartments better reflect average air concentrations and provide more reliable endpoints for discussion.

To keep the number of results to a manageable level, the various plant samples were grouped into five broad categories: forage (hay, baled hay, haylage, corn silage, alfalfa, grass and raspberry leaves), grain (corn, barley and spring wheat), TMR, fruit and fruit vegetables (apples, pears, raspberries, tomatoes, cucumber and soya meal) and root crops (mixed vegetables, potatoes, carrots, beets and garlic). Similarly, the animal products were grouped into four categories: milk, eggs, calf flesh (including calf heart) and chicken flesh (including chicken blood). Moreover, the plant and soil samples from DF8 and DF11 were combined in the analysis since the farms were so close together and the crops grown were similar. In contrast, the animal and TMR data were analysed separately because the cows had different diets. A separate analysis was also carried out for each sampling period. The average observed OBT concentrations for each of these categories are shown in Table 6.

Table 6. Average OBT concentrations (Bq L^{-1} combustion water) in the grouped samples. Where more than one sample of a given type was collected, the average and standard deviation of the measurements are listed. The numbers in brackets beside the concentrations are the number of samples in the average.

Sample Type	DF8 and DF11 combined		F27		P2
	July	September	July	September	September
Soil		20.6±1.9 (2)		32.9	552
Plants					
Forage	54.4±23.4 (5)	26.0±4.8 (8)	31.0	20.3	704±27 (2)
Grain	39.4±11.5 (2)	29.8±7.9 (3)	27.4	21.3±5.7 (2)	
Root Crops			42.0	32.9±11.1 (3)	
Fruit and fruit veg				32.6±11.1 (6)	
Animal Products					
Milk - DF8	33.9				
- DF11	21.3				
Calf flesh/heart – DF8		29.1±2.2 (2)			
– DF11		26.4±6.4 (2)			
Eggs			28.7±10.9 (3)	26.2	
Chicken flesh/blood				21.1±0.8 (2)	

Concentrations in all compartments were lower than those in air moisture, as required by specific activity concepts. The plant concentrations were higher in July than in September at all locations but the animal concentrations were the same at both sampling times, perhaps because the concentration in drinking water, which contributes significantly to the total tritium intake, varied little over time. At F27, the concentrations in vegetables and fruit were higher than in forage or grain. The standard deviations of the measured values were relatively low (< 30%) for all categories except forage at the dairy farms in July, vegetables, fruit and root crops at F27 in September and eggs at F27 in July.

Some of the variability evident in Table 6 can be reduced by normalizing the observations by the HTO concentration in air moisture, which controls concentrations in the other compartments and which varied over time and space during the study. The air moisture concentrations (in Bq L^{-1}) were derived from the air concentrations in Table 1 (in Bq m^{-3}) by dividing by 0.012 kg m^{-3}, the average absolute humidity over the growing season. The normalized results are shown in Table 7. The ratios for a given sample type incorporate data from all sampling locations and times. For rain, the ratios are based on monthly concentrations in rain and air moisture. For the other HTO endpoints, the observations are scaled by the air concentration in the month prior to sampling, the shortest interval available. For the OBT endpoints, the observations are scaled by the air concentrations averaged over the two months before sampling, to reflect the longer residence time of OBT in plants and animals.

Table 7. Observations normalized by HTO concentrations in air moisture. Results for a given sample type incorporate data from all sampling locations and times.

Sample Type	Mean	Standard Deviation	Minimum	Maximum	Number of Samples
Monthly Rain (HTO)	0.32	0.23	0.11	0.82	15
Soil (HTO)	0.33	0.03	0.29	0.36	4
Drinking water (HTO)	0.29	0.03	0.24	0.33	3
Plants (OBT)					
Forage	0.41	0.18	0.19	0.83	17
Grain	0.33	0.15	0.14	0.57	8
TMR	0.38	0.04	0.32	0.43	4
Fruit and Fruit Veg	0.30	0.10	0.19	0.50	6
Root Crops	0.30	0.09	0.16	0.38	4
Animal Products (HTO)					
Milk	-	-	-	-	0
Calf flesh/heart	0.45	0.04	0.41	0.51	4
Eggs	0.34				1
Chicken flesh/blood	0.34				1
Animal Products (OBT)					
Milk	0.28	0.06	0.22	0.35	2
Calf flesh/heart	0.39	0.07	0.28	0.46	4
Eggs	0.20	0.07	0.13	0.29	4
Chicken flesh/blood	0.19	0.01	0.19	0.20	2

The rain/air ratios show considerable variability, ranging from 0.11 to 0.82. Concentrations in rain depend strongly on the frequency with which rain falls when the plume is present and are unlikely to show stable values over averaging time as short as a month. The overall mean ratio of 0.32 falls within the range of values (0.041 – 0.44) found in other studies (Davis et al. 2002; BIOMASS 2003). The four measured soil/air ratios were all very similar at about 0.33 and also agree with the data of Davis et al. (2002) and BIOMASS (2003). The normalized drinking water concentrations show little variability, with a mean value of 0.29, but the significance of this is not clear. The drinking water samples were taken from wells and the concentrations are likely to be driven more by local hydrology than air concentrations. The normalized plant OBT concentrations varied between 0.14 and 0.83. The values for forage, grain and TMR are consistent with a plant HTO/air moisture ratio of 0.6 - 0.7, together with an isotopic discrimination factor of 0.7 in the formation of OBT. The normalized OBT concentrations for root crops, fruit and fruit vegetables, which take a lot of their tritium from the soil, tend to be lower than those for the other types of plants, which are influenced more by concentrations in air moisture. Animal OBT/air ratios ranged from 0.13 to 0.46. On average, the OBT concentrations in animal products were lower than the HTO concentrations, and lower than the OBT concentrations in the feed.

3. MODELLING APPROACHES

Eight participants submitted results for this scenario (Table 8). All participants treated the scenario as a blind test of their models and submitted results before the observed concentrations were made known to them.

Table 8. Participants in the Pickering Scenario

Participant	Affiliation	Model	Designation in text
F. Baumgärtner	Technische Universität München, Germany	BioM	TUM
R. Peterson	Lawrence Livermore National Laboratory, USA	DCART	LLNL
T. Nedveckaite	Institute of Physics, Lithuania	LIETDOS	LIET
P. Marks	GE Healthcare, U.K.	-	GE
D. Galeriu	National Institute of Physics and Nuclear Engineering – Horia Hulubei, Romania	-	IFIN
M. Saito	Safety Reassurance Academy, Japan	-	SRA
S. le Dizès-Maurel	Institut de Radioprotection et de Sûreté Nucléaire, France	TOCATTA	IRSN
D. Cutts	Food Standards Agency, UK	STAR H-3	FSA

The Pickering scenario tested models that predict tritium concentrations in a terrestrial ecosystem subject to a continuous release of HTO. It was a fairly simple scenario in the sense that releases 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. The approaches taken by the various participants to model this scenario varied widely. FSA used the STAR H-3 model, a dynamic compartment model that is formulated in terms of a series of coupled first-order differential equations. Rate constants for the transfers between compartments were derived from consideration of the hydrogen inventories of the compartments and the hydrogen fluxes between them. Predictions for the Pickering scenario, which is an equilibrium situation, were obtained from the steady-state solution to the equations. IRSN, GE, LIET and LLNL used in-house models that are well established in their respective institutions. The IRSN and GE models are similar in structure to STAR H-3, whereas LIET and LLNL are based for the most part on simple analytical equations that describe transfers between most compartments using empirically-based bulk parameters.

TUM, IFIN and SRA used less formal approaches, developing the computational tools needed to make their predictions in an *ad hoc* fashion. For the most part, these models were also analytical in structure and employed well-known empirical relationships between concentrations in the various environmental compartments. All of the modellers

grouped the plants and animals into a small number of categories to facilitate their calculations.

The TUM 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. TUM 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 TUM model, if the tritium in the hydration shells is neglected.

Although the models used by the various participants were very different in formulation, they were all based on the same pool of environmental tritium data. The rate constants used by the compartment models were derived from the same data that provided the bulk parameters used by the analytical models. Thus the differences in model structure do not necessarily imply similar differences in predictions.

The modellers used air concentrations averaged over different time intervals to drive their models. In the LLNL model, the mean air concentration from May to July was used to calculate concentrations in the samples collected in July, and the mean air concentration from May to September to calculate concentrations in the September samples. The IFIN approach was to base HTO concentrations on the air concentration in the month prior to sampling and the OBT results on the air concentration averaged over the two months before sampling. In the IRSN model, the July and September air concentrations were used to drive the predictions for the two sampling periods. The other models adopted variations on these approaches.

The FSA results are based on an absolute humidity value appropriate to UK conditions instead of the value specified in the scenario. Use of the scenario specific value for this parameter would have decreased the FSA predicted concentrations in all endpoints by approximately 1/3.

The participants also estimated the uncertainties in their predictions using very different methods. Three modellers (IRSN, LIET and LLNL) carried out a rigorous Monte Carlo uncertainty analysis using Latin Hypercube techniques to sample distributed parameters. At the opposite end of the spectrum, IFIN used expert judgment to estimate his uncertainties. Between these extremes, SRA carried out an analytical analysis, on the

assumption that the uncertainty in each input parameter was ±20%. TUM, GE and FSA did not submit uncertainty estimates.

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 C.

4. COMPARISON OF PREDICTIONS AND OBSERVATIONS

4.1 Soil Water

Predictions for the HTO concentration in soil water at DF8 and DF11 combined are compared with the observation in Figure 2. Five of the six models that submitted predictions for this endpoint produced results in good agreement with the observation even though they were all very different in structure. LLNL assumed the soil water concentration equalled 30% of the air moisture concentration, following the recommendation of BIOMASS (2003). IFIN assumed that the tritium in soil arose primarily from washout and set the soil water concentration equal to the sum of the concentration in rain plus 10% of the concentration in air moisture. SRA used a more complex analytical equation that described the balance between average tritium sources (wet and dry deposition) and sinks (infiltration, plant uptake and re-emission) in the root zone. The FSA and IRSN models are similar to this since, at steady state, the coupled differential equations on which they are based lead to solutions that are essentially a balance between sources and sinks.

The predictions of these five models for soil water concentrations were as good or better at F27 and P2 as they were at the dairy farms. Thus, good model performance for this data set can be achieved with models of very different complexity. In contrast, the predictions of the LIET model overestimated the observed soil water concentrations by about a factor of two at all sites. This model obtained the soil concentrations by balancing gains and losses in a two-compartment model of air and soil. The soil concentration was expressed in terms of the concentration in rain, the soil water content, the average rainfall rate, the depth of the root zone and the rate constant for losses from soil due to evapotranspiration, infiltration and runoff. The overprediction may have been due to an inappropriate choice of values for those parameters that were not defined in the scenario description.

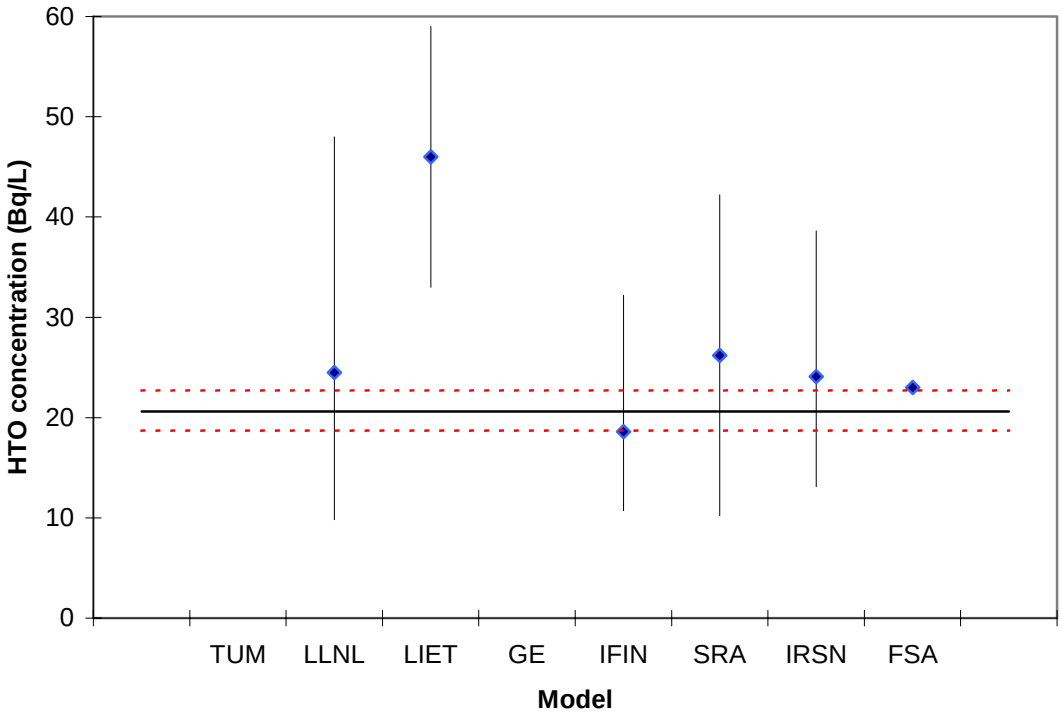

Figure 2. HTO concentration in soil water for the September sampling period at DF8 and DF11 combined. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines. FSA did not estimate uncertainties and TUM and GE did not submit results for this endpoint.

The 95% confidence intervals shown in Fig. 2 are fairly consistent from model to model, despite the different approaches taken by the participants in estimating their uncertainties. The confidence interval for LIET is clearly an underestimate since the prediction does not agree with the observation even when uncertainties are taken into account. The confidence intervals for the other endpoints were similar and will be discussed further in Section 5.

4.2 Forage

Predictions of the OBT concentration in forage crops at DF8 and DF11 combined for the September sampling period are compared with the observation in Figure 3. The GE result, which was reported in Bq kg^{-1} fresh weight, was converted to Bq L^{-1} water equivalent assuming a water fraction of 0.75 for fresh forage and a water equivalent

factor of 0.59. With the exception of TUM, the scatter in the predictions was relatively small. However, all models overestimated the observed concentration, by up to a factor of 3 in the case of LIET and GE, and by a factor of 2.3 on average. The results of four models (LIET, IFIN, SRA and IRSN) marginally agreed with the data when uncertainties were taken into account. The TUM model underestimated the observation, but this was expected since this model predicts the concentration of buried tritium rather than fixed OBT. Similar results were obtained for DF8 and DF11 in July, although the degree of overprediction was not as large, and the results of all five models that estimated uncertainties agreed with the observation when the uncertainties were taken into account. However, the better agreement in July could be primarily a result of anomalously large measured concentrations in hay and haylage at DF8 rather than improved model performance. All of the models overestimated the OBT concentrations in grass at F27 by a factor of at least 3, and in grass and raspberry leaves at P2 by a factor of 2 on average.

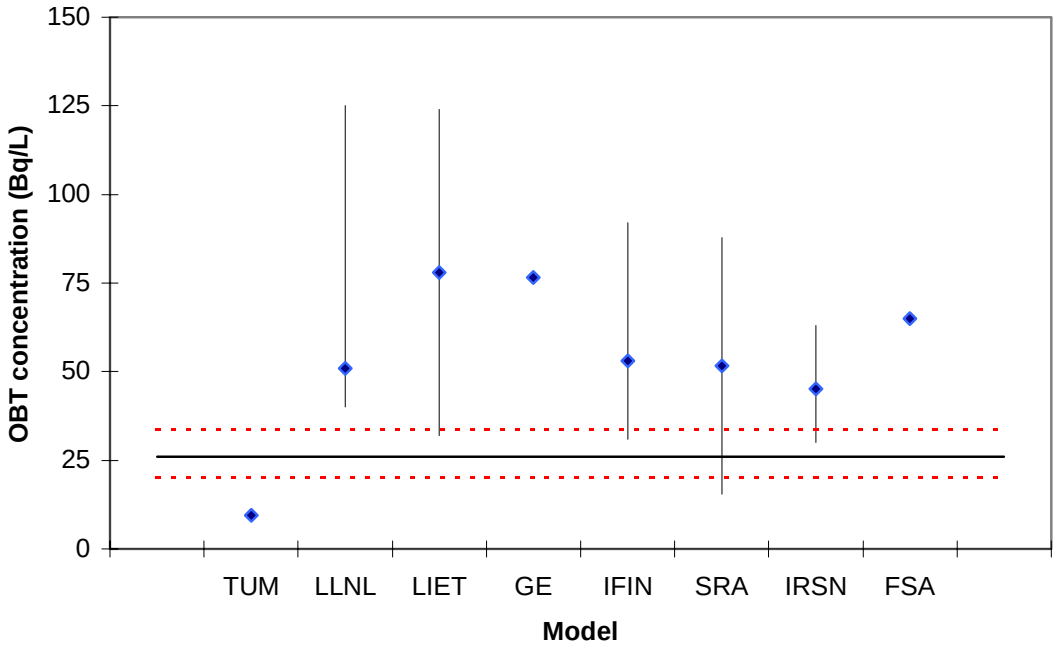

Figure 3. OBT concentration in forage crops for the September sampling period at DF8 and DF11 combined. TUM, GE and FSA did not estimate uncertainties for this endpoint.

OBT concentrations depend on the HTO concentration in the plant leaves and the rate at which that HTO is converted to OBT. The reasons for the misprediction of OBT concentrations evident in Fig. 3 must be sought in these processes and they way they were modelled. The various participants determined the HTO concentration in plants in very different ways. Six models (FSA, IRSN, SRA, GE, LIET and LLNL) explicitly took into account the transfers of tritium to the plant from air and soil. FSA, GE and IRSN did this by specifying appropriate rate constants for use in their numerical models and calculating plant HTO concentrations at steady state. SRA used an analytical

equation that balanced uptake and loss, with the roles of rainfall and air-plant transfer expressed explicitly:

$$C_{pw} = \alpha \left[\frac{C_a + I_w C_{sw} r}{\rho_s + \alpha\, I_w r} \right], \qquad (1)$$

where C_{pw} is the HTO concentration in plant water,
$\alpha = 1.1$ is the ratio of the vapour pressure for water vapour to that of HTO,
C_a is the HTO concentration in air,
I_w is the average rainfall intensity,
C_{sw} is the HTO concentration in soil water,
ρ_s is the saturated vapour density of the air, and
r (= 67 s m^{-1}) is the exchange resistance for HTO and water between the plant leaf and the atmosphere.

LLNL and IFIN calculated the plant HTO concentration using Murphy's (1984) analytical model, which distinguishes the contributions of air moisture and soil water to the HTO concentration in the plants:

$$C_{pw} = \alpha\, [RH\, C_{am} + (1\text{-}RH)\, C_{sw}], \qquad (2)$$

where RH is the relative humidity and C_{am} is the HTO concentration in air moisture. LIET used an equation similar to Eq. (2) but with a slightly larger contribution from the soil. The remaining model (TUM) took a more empirical approach, assuming that C_{pw} was equal to the mean of the HTO concentration in drinking water and in rainfall (averaged over the 2-3 months prior to sampling); where the drinking water concentration was not available in July, C_{pw} was set equal to the average concentration in rain.

The predictions of the eight models for the HTO concentration in plant water for forage crops at the dairy farms are shown in Table 9. The results vary over a factor of more than 2 for July and more than 3 for September. The scatter is about a factor of two even for the six models that are theoretically based. Also shown in Table 9 are the plant concentrations normalized by the average air moisture concentrations in the month prior to sampling (103 and 64.6 Bq L^{-1} for the July and September sampling periods respectively). Some of the predictions show a plant/air ratio greater than 1, and most have a ratio greater than 0.65, the long-term average value observed in forage crops (Peterson and Davis 2002), but this could easily be due to the mismatch in averaging times for air and plant. The HTO predictions show a pattern similar to that evident in Fig. 3 and explain most of the variability in the OBT results. The high plant/air ratios are likely responsible for some of the overprediction. Unfortunately, long-term average HTO measurements in plant water are not available to help identify the best predictions.

Table 9. Predicted HTO concentrations in plant water for forage crops at the dairy farms

Model	HTO Concentration			
	July		September	
	Plant (Bq L^{-1})	Plant/Air	Plant (Bq L^{-1})	Plant/Air
TUM	47	0.46	24.8	0.38
LLNL	85.5	0.83	72.9	1.13
LIET	97	0.94	97	1.50
GE	100	0.97	71	1.10
IFIN	74	0.72	53	0.82
SRA	54.9	0.53	44.9	0.70
IRSN	-	-	50.2	0.78
FSA	78	0.76	56.0	0.87

The other processes controlling OBT concentration are the rates of OBT formation and loss in the plant. The numerical models (FSA, IRSN and GE) accounted for these processes directly. In the analytical and empirical models, the OBT concentration was calculated as a fixed fraction of the HTO concentration. The TUM model calculated the concentration of buried tritium rather than OBT itself using a two-step exchange process that accounted for the number of exchangeable hydrogen positions in the carbohydrates and proteins of the plant in question.

The OBT/HTO ratios for each model are shown in Table 10. All but one of the ratios are high compared to observed ratios in the field (Peterson and Davis 2002), which tend to scatter about 0.7. Three of the models, including two of the numerical models, predict OBT concentrations larger than the corresponding HTO concentrations. The value used by IFIN was chosen to be deliberately conservative. These large values explain part of the general overprediction of OBT concentrations in the forage crops.

Table 10. OBT/HTO ratios in forage crops at DF8 and DF11

Model	OBT/HTO ratio
LLNL	0.7
LIET	0.8
GE	1.1
IFIN	1.0
SRA	1.1
IRSN	0.9
FSA	1.2

No data are given in Table 10 for the TUM model, which calculates the concentration of buried tritium rather than fixed OBT. The predictions for buried tritium lay between one

third and one half of the observed OBT concentrations. If these predictions are correct, buried tritium makes up a significant proportion of what is traditionally called OBT.

4.3 Grain, Fruit Vegetables, Fruit and Root Crops

Two modellers (IRSN and FSA) assumed that the HTO concentration was the same in the edible portions of grain, fruit vegetables, root crops and fruit as it was in forage. The other modellers reduced the HTO concentrations in these plants to account for the fact that they draw more of their tritium from soil water than the forage crops do. However, all of the modellers assumed that HTO was taken up by the leaves of all plant types in the same way, that OBT was formed in the leaves by photosynthesis, and that the OBT was translocated to the edible portion of the plant without change in concentration. Thus, each participant predicted the same OBT concentration in all crops sampled at the same time and place. Leaving the TUM results aside for the moment, all of the models overestimated the OBT concentrations in all crop types at all sampling sites and times. The degree of overprediction for the various crops is shown in Table 11 in terms of the mean ratio of predictions to observations (the mean P/O ratio). The TUM and GE results were not included in these factors, since TUM did not calculate traditional fixed OBT *per se* and the very high GE predictions suggest a mistake may have been made. There is a tendency for the ratios to be higher at F27 than elsewhere. This conclusion cannot be stated definitively for forage and grain since the results are based on one or two samples only and the measured concentrations may be unreliable. But the overprediction for fruit, fruit vegetables and root crops must be accepted as real and suggests that the models are not performing as well for these crops as for forage and grain. The results for fruit and fruit vegetables measured at F27 in September are shown in Fig. 4, where the mean overprediction was 2.6.

Table 11. Average factor by which the predictions overestimated the observations for OBT in plants

Crop type	Site	Month	Mean P/O ratio
Forage	DF8 and DF11	July	1.4
		September	2.3
	F27	July	3.4
		September	4.5
	P2	September	1.9
Grain	DF8 and DF11	July	1.8
		September	1.9
	F27	July	3.3
		September	4.0
Root Crops	F27	September	2.6
Fruit and Fruit Vegetables		September	2.6

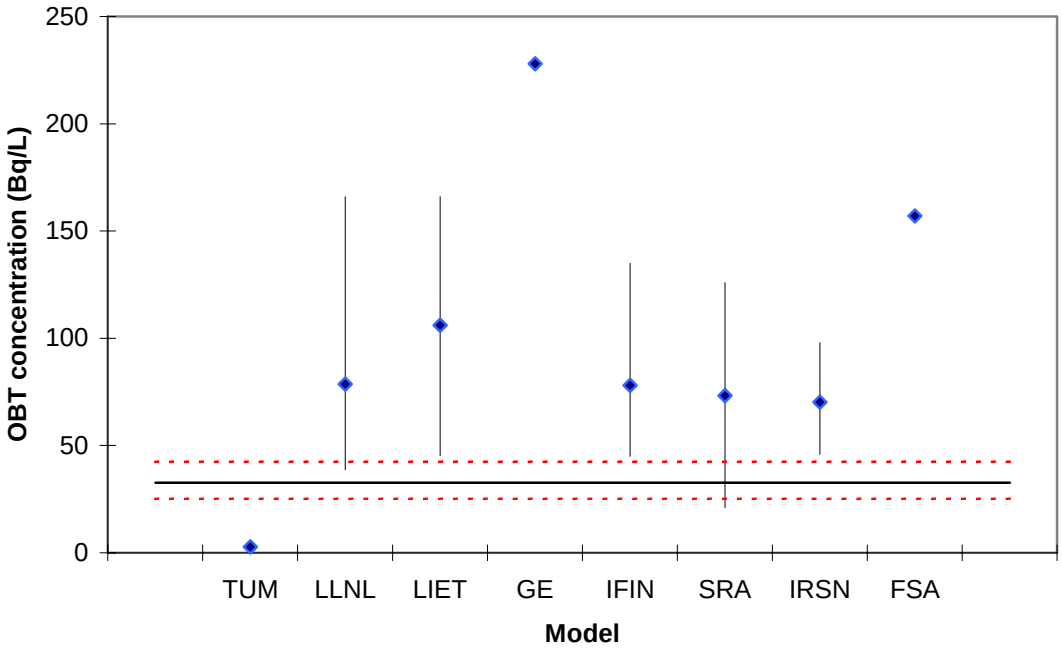

Figure 4. OBT concentration in fruit and fruit vegetables for the September sampling period at F27. TUM, GE and FSA did not estimate uncertainties for this endpoint.

4.4 Total Mixed Ration (TMR)

The calculation of TMR concentrations required special consideration for two reasons: (i) not all of the components of TMR were contaminated and (ii) most of the TMR fed to the cows in 2002 was grown in 2001. The LLNL, IFIN and SRA models took both of these factors into account, calculating concentrations in the various components of the 2001 TMR using the air concentrations measured in 2001, and forming the TMR concentration itself from an average of the component concentrations weighted by their fractional contribution to the total make-up of the TMR (with the uncontaminated components assumed to have background tritium levels). IRSN accounted for the higher air concentrations in 2001 but not the uncontaminated portion of the TMR; LIET accounted for the uncontaminated portion but not the higher air concentrations. GE took neither of these factors into account but instead set the TMR concentration equal to the concentration of the forage crops (on a fresh weight basis). FSA did not submit predictions for TMR.

Predictions for the OBT concentration in the TMR sample collected at DF11 in July (which was composed of crops harvested in 2001) are shown in Fig. 5. Similar results were obtained for DF8 and the September sampling period. All of the models overestimate the observed concentration, although not as severely as some of the other endpoints. Predictions of five of the six models agree with the observation when uncertainties are taken into account.

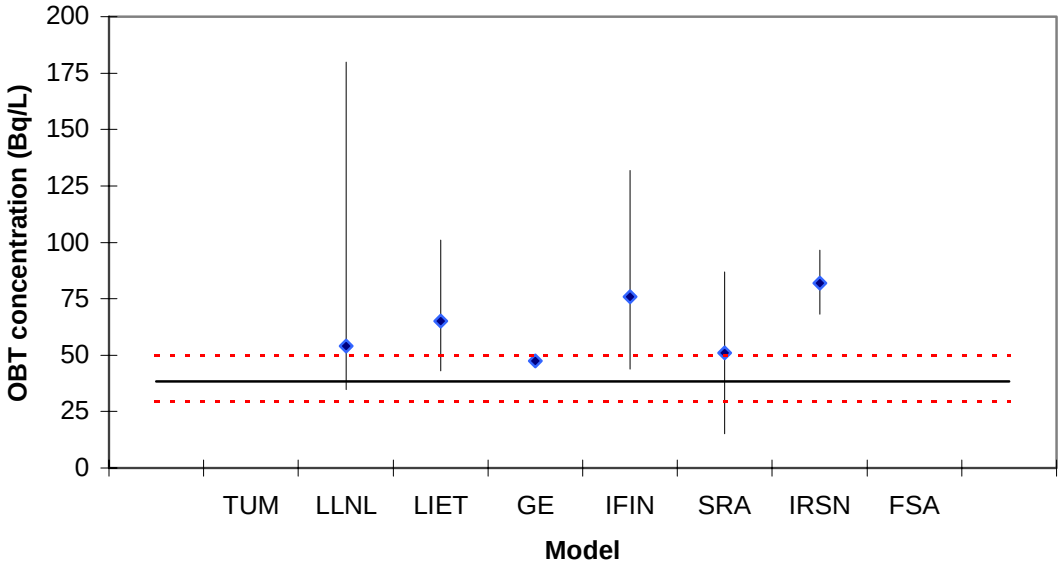

Figure 5. OBT concentration in the TMR sample collected in July at DF11. TUM and FSA did not submit predictions for this endpoint.

4.5 Milk and Beef

4.5.1 HTO Concentrations: Predictions of the average HTO concentration in calf flesh and heart for the samples taken at DF8 in September are compared with the observation in Fig. 6. With the exception of FSA, the predictions ranged over less than a factor of two and all agreed with the observed value when uncertainties were taken into account. Similarly good agreement was obtained for the HTO concentrations in calf flesh and heart at DF11 in September, even though the diet of the cows was not well known at that site. The assumptions made by the various modellers regarding the ingestion rate of the cows at DF11 are shown in Table 12. The differences in the assumed value would have contributed to the variability in the predicted concentrations.

Unfortunately, HTO concentrations were not measured in the milk samples so the predictions could not be compared with observations. But the predictions of most of the models show the same relatively small scatter evident in Fig. 6 at both DF8 and DF11. When FSA, which appears to be an outlier, was left out of the calculations, the mean predicted HTO concentration in milk was about 30 Bq L^{-1} at both sites, with a standard deviation of less than 30%.

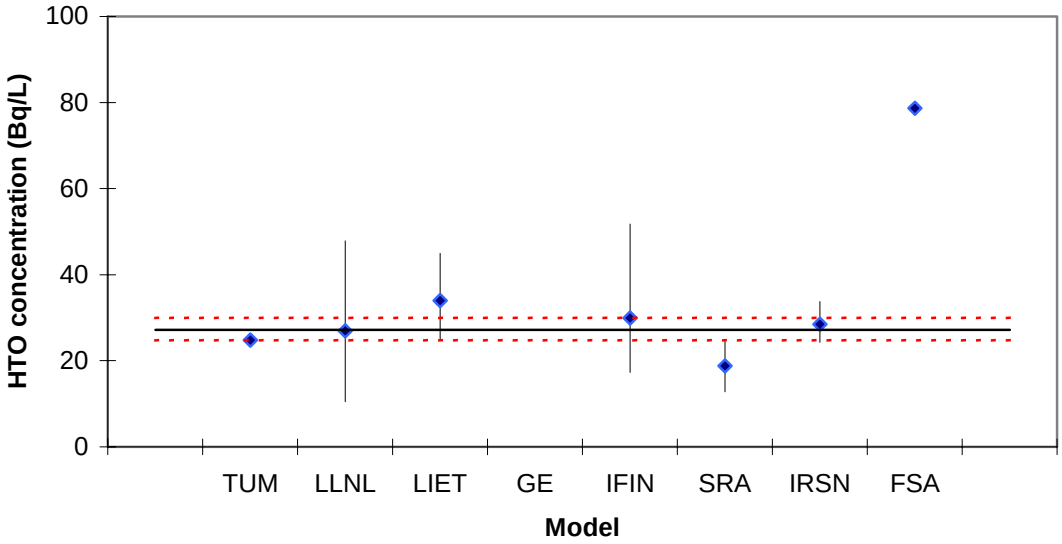

Figure 6. Average HTO concentration in calf flesh and heart at DF8 in September. GE did not submit a prediction for this endpoint and TUM and FSA did not estimate uncertainties.

Table 12. Values adopted by the various modelers for food and drinking water ingestion rates

Model	Ingestion rate of cows at DF11 (kg dry d^{-1})	Ingestion rate of chickens at F27 (kg dry d^{-1})	Drinking water ingestion rates (L d^{-1})	
			Cows	Chickens
LLNL	16.4	0.139	80	0.29
LIET	14	0.1	35	0.2
IFIN	19	0.2	70	0.3
SRA	10	0.1	90	0.2
IRSN	10	0.2	75	0.3
FSA	115 (fresh wt)	0.5 (fresh wt)	60	0.2

The agreement in the predicted HTO concentrations was achieved despite the fact that the models used by the various participants were quite different. In their numerical models, FSA and IRSN specified rate constants that described the uptake of tritium by the animal through inhalation and ingestion, and losses due to elimination, and solved for the concentrations at steady state. LLNL assumed that the animal HTO concentration was equal to the average concentration of the water pools accessed by the animal (plant water, plant organic matter, drinking water and inhalation/skin absorption), weighted by the fraction that each pool contributed to the total water intake. IFIN used a model based on the metabolism of hydrogen and carbon in the body to derive transfer parameters specific

to the animal in question and its diet. SRA used the experimental data of Kirchmann et al. [1977, 1985] to derive the tritium specific activity in animal products given the specific activity in the diet and the drinking water. LIET expressed the animal concentrations in terms of the fraction of daily tritium intake that appears in the animal product, with separate values for transfer from HTO in food to HTO in animal product, from OBT in food to HTO in animal product, from HTO in food to OBT in animal product and from OBT in food to OBT in animal product. TUM assumed that the animal concentration was equal to the mean of the HTO concentration in drinking water and in rainfall averaged over the 2-3 months prior to sampling. GE did not calculate animal concentrations.

The similarity in predictions despite the divergence in model structure can be attributed in part to the fact that drinking water is a major contributor to tritium body burden and that drinking water concentrations were provided with the scenario. The ingestion rates assumed by the modelers (Table 12) imply that drinking water contributed between 50 and 80% to the total tritium body burden of the cows. Thus, knowing the tritium concentration in drinking water helped to damp the effect of the overprediction of food concentrations.

4.5.2 OBT Concentrations: Predictions of the average OBT concentration in calf flesh and heart for the samples taken at DF8 in September are compared with the observation in Fig. 7. The agreement between predictions and observations is worse than it was for HTO. The predictions show greater scatter, ranging over a factor of 10, and only three agree with the observed value when uncertainties are taken into account. Most of the models overpredict the observation, with a mean P/O ratio of 1.6. Similar results were obtained for the OBT concentrations in calf flesh and heart at DF11, where the mean P/O ratio increased to 2. Results for milk were also similar, with considerable scatter in predictions at both sites and mean P/O ratios of 1.2 and 2.3 at DF8 and DF11, respectively.

Four participants considered HTO and OBT to be coupled within the cow and solved for the concentrations of the two species simultaneously using the same model. Thus the numerical models of FSA and IRSN, the metabolic model used by IFIN and the transfer parameter model of LIET returned OBT concentrations as well as HTO. SRA used the empirical data of Kirchman et al. [1977, 1985] for both HTO and OBT. LLNL set the OBT concentration equal to the HTO concentration and TUM assumed an exchange process model to calculate the concentration of buried tritium. The differences in these models and their parameter values resulted in the scatter evident in Fig. 7. Differences in assumptions for the food ingestion rate at DF11 and in the water ingestion rates at both sites (Table 12) would also have contributed to the variability in the predicted concentrations.

The models differed in their predictions of the ratio of OBT to HTO concentrations in milk and calf flesh. One model (RSA) produced an OBT/HTO ratio of about 0.6. Two other models (LLNL and FSA) predicted a ratio close to 1. In the remaining models (LIET, IFIN and IRSN), the OBT concentrations exceeded the HTO concentrations, by a

factor of 2 on average. In fact, the data show that the HTO and OBT concentrations in calf flesh are about the same. This observation may be specific to the conditions of this scenario and not generally applicable. The primary source of HTO for the cows was drinking water whereas the main source of OBT was TMR, and concentrations in these two sources were essentially independent.

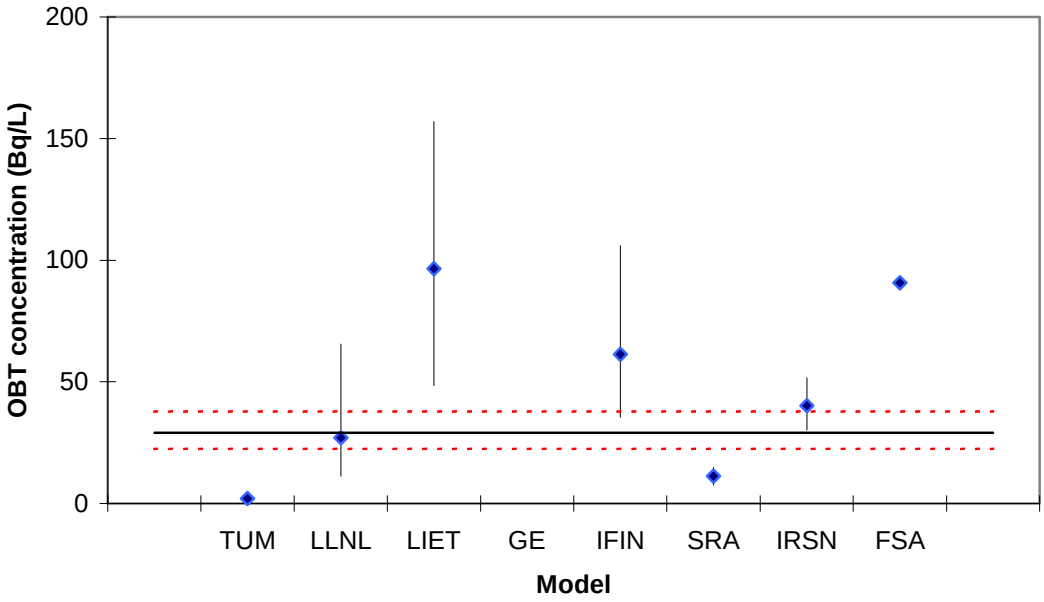

Figure 7. Average OBT concentration in calf flesh and heart at DF8 in September. GE did not submit a prediction for this endpoint and TUM and FSA did not estimate uncertainties.

The data show that the OBT concentration in milk or flesh in July was about 30% lower than the concentration of OBT in TMR grown in 2001. In September, the situation was reversed, with the OBT concentration in milk or flesh about 20% greater than that in TMR. The latter finding is surprising since much of the OBT ingested by the cow is expected to be converted to HTO during digestion, and little of the HTO ingested is converted to OBT. Most modelers predicted animal concentrations lower than TMR concentrations, by factors that ranged from 0.25 for SRA to 0.8 for IFIN and IRSN. In contrast, the results for LIET and FSA showed animal concentrations as much as 50% greater than those in TMR.

With two exceptions, the models predicted that the OBT concentrations in flesh and milk were about the same. The exceptions were LIET and FSA, which predicted flesh concentrations greater or less than those in milk depending on the site and the time of sampling. Observations are not available to test these predictions since milk and flesh were never sampled at the same time.

4.6 Chicken and Eggs

4.6.1 HTO Concentrations: Predictions of the HTO concentration in eggs for the sample taken at F27 in September are compared with the observation in Fig. 8. The performance of the models is not as good for eggs as it was for milk or calf flesh. The predictions show considerable scatter, with only three agreeing with the observation when uncertainties are taken into account. Three of the results overestimated the observation by factors ranging from 2 to 4. Similar results were obtained for the HTO concentrations in chicken blood in September. The scatter was much the same for the predicted concentrations in eggs in July, although in this case no observation was available for comparison. The participants used the same models for chickens and eggs as they did for milk and calf flesh, so the poorer performance here must be due to the parameter values used in the models. In particular, the feed and water ingestion rates for the chickens were not known and the modellers made very different assumptions about their values (Table 12), which would have contributed to the variability in the predicted concentrations. Also, the models assume all drinking water was contaminated, when in reality the chickens may have drawn their water from uncontaminated sources.

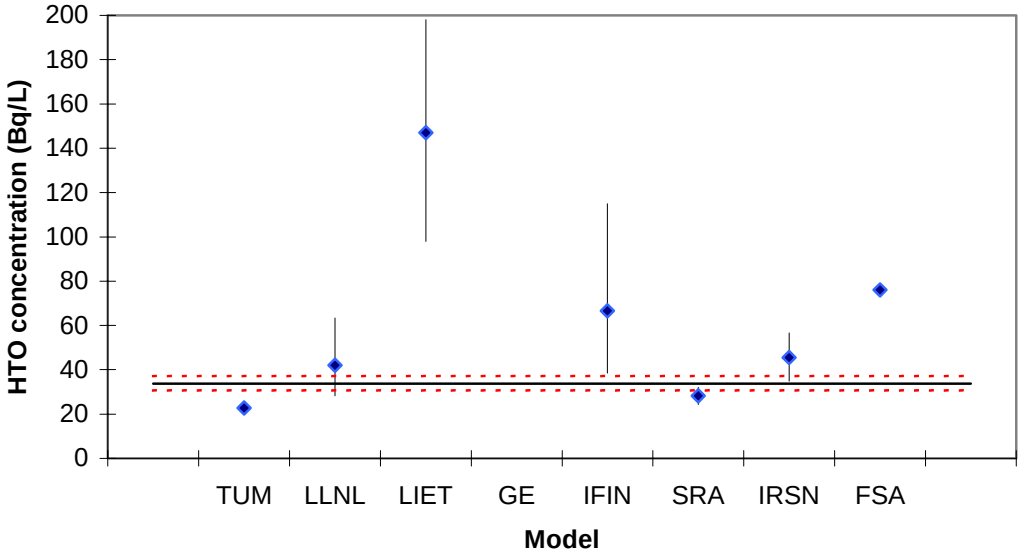

Figure 8. HTO concentration in eggs at F27 in September. GE did not submit a prediction for this endpoint and TUM and FSA did not estimate uncertainties.

4.6.2 OBT Concentrations: Predictions of the OBT concentration in eggs for the sample taken at F27 in September are compared with the observation in Fig. 9. The scatter among the models was less than it was for HTO, but the level of agreement between predictions and observations was worse, with all of the models apart from TUM overpredicting the measured value, by a factor of 3.2 on average. Only the LLNL model

agreed with the observation when uncertainties were taken into account. Similar results were obtained for the OBT concentration in eggs in July. Results were worse for chicken blood and flesh in September, where the mean P/O ratio increased to 4.5.

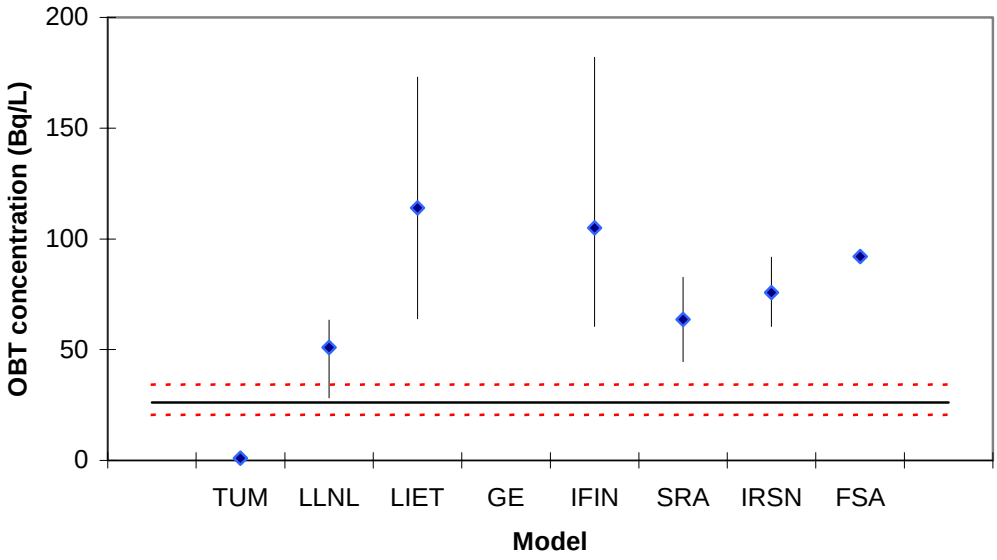

Figure 9. OBT concentration in eggs at F27 in September. GE did not submit a prediction for this endpoint and TUM and FSA did not estimate uncertainties.

With one exception, the models consistently predicted higher OBT than HTO concentrations in eggs and blood, with the OBT/HTO ratio varying from 1.2 to 2.5. The exception was LIET, which predicted an OBT/HTO ratio of 0.47 for eggs in July, 0.78 for eggs in September and 1.04 for blood in September. In fact, the data show that the OBT concentration was less than the HTO concentrations, with an OBT/HTO ratio of 0.78 for eggs and 0.63 for blood. As was the case for cows, this observation may not be generally applicable outside of this scenario.

The data show that the OBT concentration in eggs and chicken flesh in September was about the same as the average OBT concentration in the feed eaten by the chickens. Most of the modelers (LIET, SRA, IRSN and FSA) reproduced this observation. In contrast, LLNL predicted an animal/feed ratio of 0.57 and IFIN a ratio of 1.4 for eggs and 1.8 for flesh.

For all models, the predicted HTO concentrations in eggs were essentially identical to the HTO concentrations in chicken flesh and blood, in agreement with the observation. With two exceptions, the models also predicted that the OBT concentrations in flesh and eggs were about the same, a conclusion again supported by the observations. The exceptions were LIET and IFIN, which both predicted flesh concentrations about 30% greater than those in eggs.

5. DISCUSSION AND CONCLUSIONS

The Pickering scenario provided a good test of models that predict tritium concentrations in the various compartments of an agricultural ecosystem at steady state. Reliable estimates of HTO concentrations were available in air moisture, precipitation and drinking water as input to the models. Most of the information required to evaluate the animal pathways was available without the need for the assumptions that usually have to be made about diet or the fraction of feed that is contaminated. On the other hand, the scenario was not ideal since some information on ingestion rates was incomplete or missing, and this contributed to the differences between predictions and observations. But many real assessments must be carried out with even less information and difficulties of this sort must be expected in practice.

The models used by the participants in their calculations varied from numerical dynamic compartment models (solved for steady-state conditions) to simple analytical models based on empirical data. Similarly, different parameters appeared in the different models, although all were based on the same pool of environmental tritium data. For these reasons, it was often difficult to explain why one model produced a different result than another, or why a specific model result differed from the corresponding observation.

Despite their differences, all models but one performed well for HTO in soil, predicting concentrations that agreed with each other and with the observations when uncertainties were taken into account. In contrast, all of the models significantly overestimated the OBT concentrations in plants, by an average factor of 1.9 at the dairy farms and 3.4 at F27. This appears to be due in part to overprediction of the concentration of HTO in the plant leaves, where OBT is formed by photosynthesis. For most models, the ratio of predicted HTO concentration in plant leaves to observed HTO concentration in air moisture was substantially larger than the value of 0.65 that has been observed in other studies. Additionally, the models appear to underestimate the effect of isotopic discrimination in OBT formation. Most of the predicted OBT/HTO ratios for the plant leaves were larger than the value of 0.7 observed elsewhere.

These two factors alone could explain overestimates of as much as a factor of two in the predicted OBT concentrations for several of the models, and resolve the differences between predictions and observations for forage and grain at the dairy farms. Additional reasons must be found to explain the more severe overpredictions at F27. One possibility may lie in the fact that most of the samples taken at this site were root crops, fruit and fruit vegetables. OBT that appears in the edible parts of these plants must be translocated from the leaves where it is formed, and a reduction in concentration may occur during the translocation process. This cannot explain the large overestimates for forage crops at F27 but the observed values for these plants may not be reliable since they were based on one or two samples only.

A second explanation may lie with the air concentrations provided as part of the scenario description. The measured concentrations at F27 were lower than those observed at the dairy farms. This was thought unlikely since the wind blows with equal frequency toward F27 and the dairy farms, and F27 is closer to the reactors. Moreover, the measurements were made with passive samplers, for which the uncertainty is large. It was therefore assumed that the measurements were in error, and, as noted in Section 1, they were replaced with predictions from a sector-averaged Gaussian plume model, which produced results in good agreement with the observed air concentrations at P2 and the dairy farms. If the measured concentrations were indeed correct and had been used in the models, the predicted plant concentrations would have been lower by a factor of 2, removing a lot of the discrepancy between predictions and observations at F27. A quantitative assessment of the air concentrations used to drive the models is given in Appendix A, based on data that became available only after work on the scenario had been finalized.

No conclusions could be drawn about the ability of the models to predict HTO concentrations in plants. HTO is very mobile in plants and the observed concentrations reflect the air concentrations in the hour or two before sampling. It is unlikely that this will match the long-term average air concentration used to drive the models, with the result that predicted and observed values cannot necessarily be expected to agree.

Most of the models predicted HTO concentrations in milk and calf flesh that were in good agreement with the observations. This may be due in large part to the importance of drinking water concentrations, which were provided in the scenario description, to the body burden of the animal. Model performance was not as good for OBT, which was overestimated in most cases. The models did not do as well for eggs and chickens as for milk and calf flesh, partly because the concentrations in chicken feed were overestimated to a greater extent than in cow feed and partly because the ingestion rates of feed and drinking water were not known for the chickens. Most of the models did not correctly reproduce the observed OBT/HTO ratio in the animals, and some predicted higher OBT concentrations in animals than in their feed, which seems unlikely in reality. Most models predicted that concentrations in milk were similar to concentrations in calf flesh, and that concentrations in eggs were similar to concentrations in chicken flesh, in agreement with the observations.

No one model stood out as generating predictions superior to the others for HTO concentrations in soil water or OBT concentrations in plants. Generally speaking, the level of agreement between predictions and observations was about the same for the numerical models as for the analytical models, although the numerical models tended to be responsible for all of the very high predictions. All of the models were satisfactory for HTO concentration in milk and calf flesh. However, the LLNL model stood out as the only model that reproduced the observed concentrations in all of the animal endpoints within the estimated uncertainties. The IRSN also did well in this regard. Despite the fact that some models predicted OBT/HTO ratios greater than one for some plants, and OBT concentrations in animals that exceeded the OBT concentration in their feed, there is no evidence in the Pickering data of tritium bioaccumulation in the terrestrial pathways.

The results of the TUM model, which calculates the concentration of buried tritium rather than the tritium traditionally considered to be organically bound, were lower than those of the other models for the OBT endpoints. The TUM predictions made up a significant proportion (40%) of the measured OBT concentrations only for forage; for fruit, fruit vegetables, calf flesh, calf heart and eggs, buried tritium made up less than 5% of the measured concentration. The results of the TUM model indicate that the formation of buried tritium is better modeled as a two-step exchange process rather than as a one-step process.

The uncertainties estimated by the various participants differed somewhat from model to model and endpoint to endpoint, but were roughly consistent with a confidence interval (97.5^{th} percentile divided by the 2.5^{th} percentile) of a factor 3. In general, the modellers estimated higher uncertainties for OBT concentrations than for HTO, which is reasonable given that the uncertainties in OBT include those for HTO plus additional ones specific to OBT itself. The uncertainty estimates for the animal endpoints were generally lower than those for plants, which is justified based on model performance for HTO in milk and calf flesh but not for HTO in eggs and chicken flesh or OBT in any animal product.

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APPENDIX A

Model Performance as a Function of Air Concentration Averaging Time

Most participants in the Pickering scenario overestimated OBT concentrations in most plant and animal products by a factor ranging from 2-5. The overpredictions were attributed to a number of factors, including a conservative bias in the model for HTO concentration in plants and the use of high values for the isotopic discrimination factor. Another possible explanation is investigated here, namely that the air concentrations used to drive the models were not the most appropriate.

The air concentrations given in the scenario description for sampling site P2 were based on measurements of the monthly average concentrations from an active air sampler, which were considered reliable. However, at the other sampling locations (DF8, DF11 and F27), air concentrations were available only as annual averages from passive diffusion samplers. These observations showed some unexpected features. Concentrations at DF8 and DF11 differed by 60% despite the fact that these two farms are located close together. Similarly, the observed concentration at F27, which is closer to PNGS than either of the dairy farms and experiences comparable meteorology, was lower than the concentration at DF8 or DF11. Finally, a comparison carried out by the utility showed that the concentrations measured by a number of passive samplers at the same location differed by a factor of 2 on average.

For these reasons, the observed air concentrations at DF8, DF11 and F27 were deemed untrustworthy and were replaced with the predictions of a sector-averaged atmospheric dispersion model that produced concentrations in good agreement with the observations at P2 and the dairy farms. The model was used to predict annual average concentrations because, at the time, annual average meteorological data were all that were available. The monthly concentrations at DF8, DF11 and F27 were deduced from the observed monthly variation at P2. The uncertainties in these concentrations, which were the concentrations given in the scenario description, were therefore high. The values averaged over the two months prior to the September sampling period are shown in Table A1. This averaging time was chosen to reflect the mean conditions under which the OBT observed in September was formed, given that OBT has a biological half-life of a few weeks in plants and animals.

The opportunity to construct more accurate air concentrations arose when monthly meteorological data became available shortly after work on the scenario was finalized. The atmospheric dispersion model was used with these data to generate monthly average air concentrations for DF8, DF11 and F27. The predictions for DF8 and DF11 were found to be 20% lower than the concentrations initially supplied to the modellers, and 35% lower at F27 (Table A1). These reductions resulted in improved model performance at all sampling sites, but still left a large gap between predictions and observations.

Table A1. Air concentrations (Bq m^{-3}) averaged over the period 2002 July 18 - September 17. All values include a background of 0.19 Bq m^{-3}.

	DF8 and DF11	F27
Values provided in the scenario description	0.84	1.30
Values calculated from monthly meteorological data	0.68	0.84
Values calculated from monthly meteorological data (daylight hours only)	0.29	0.26

Model performance was investigated for one further averaging time. HTO transfer between air and plant, and OBT formation, occur more rapidly during the day than at night, suggesting that daylight air concentrations may be more relevant in determining plant tritium concentrations than 24-hour concentrations. Accordingly, the dispersion model was used to calculate daylight air concentrations over the period July 18 to September 17. These were found to be a factor 2-3 lower than the 24-hour averages (Table A1) because of the prevalence of unstable conditions during the day and stable conditions at night. Since plant concentrations are directly proportional to air concentrations, OBT predictions for daylight conditions were found by multiplying the initial result for each model by the ratio of the daylight air concentration to the concentration provided in the scenario description. The results are shown in Figures A1 (for OBT concentrations in forage crops at DF8 and DF11 combined) and A2 (for OBT concentrations in fruit and fruit vegetables at F27). In each case, the figure showing the original results for each model is repeated from the main text, followed by the results obtained for daylight air concentrations. The use of daylight concentrations dramatically improves the performance of the models, with essentially all of the predictions agreeing with observations when uncertainties are taken into account.

The predicted OBT concentrations in animal products corresponding to daylight air concentrations could not be found using the simple scaling applied above to plants since animal concentrations are not directly proportional to air concentrations: drinking water provides an additional, independent intake route. To estimate the animal concentrations without re-running all the models, the LLNL model was used to determine the ratio of the OBT concentration calculated from daylight air concentrations to the OBT concentration determined from the concentrations given in the scenario description. This ratio was then applied to all model results (Figures A3 and A4). As was the case for plants, the use of daylight air concentrations brings the predictions into much better agreement with the observations, although some variability is observed from model to model, and the concentration in eggs is still overestimated by all models except TUM.

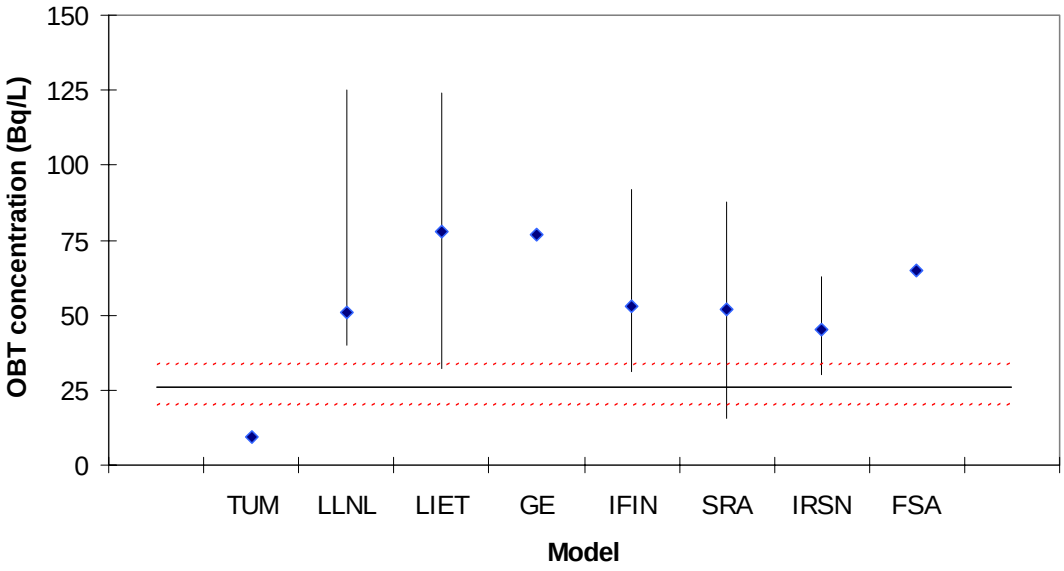

Figure A1a. OBT concentrations in forage crops for the September sampling period at DF8 and DF11 combined, predicted using the air concentrations given in the scenario description. The model predictions are shown as solid diamonds with the vertical lines representing 95% confidence intervals as estimated by the modelers. The solid horizontal line is the observation with the 95% confidence interval indicated by the dashed lines.

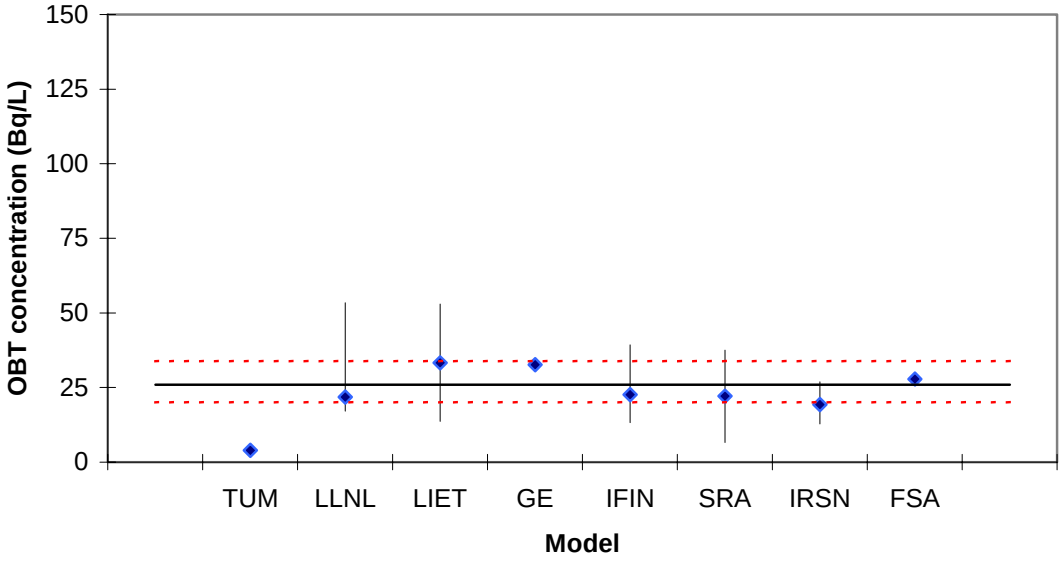

Figure A1b. As in Fig. A1a but predictions were obtained using air concentrations calculated from monthly meteorological data (daylight hours only)

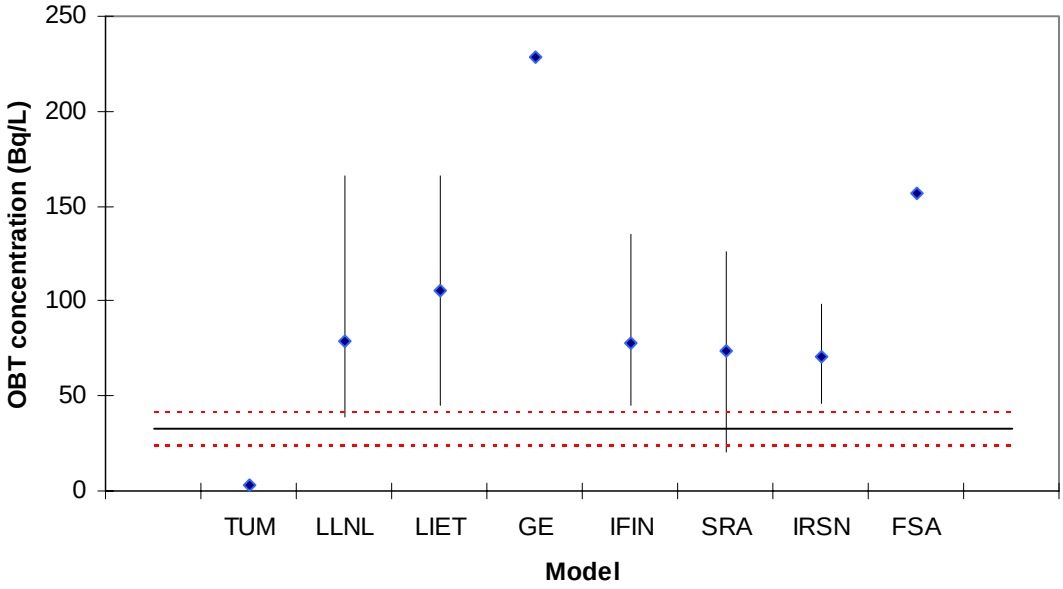

Figure A2a. OBT concentrations in fruit and fruit vegetables for the September sampling period at F27, predicted using the air concentrations given in the scenario description.

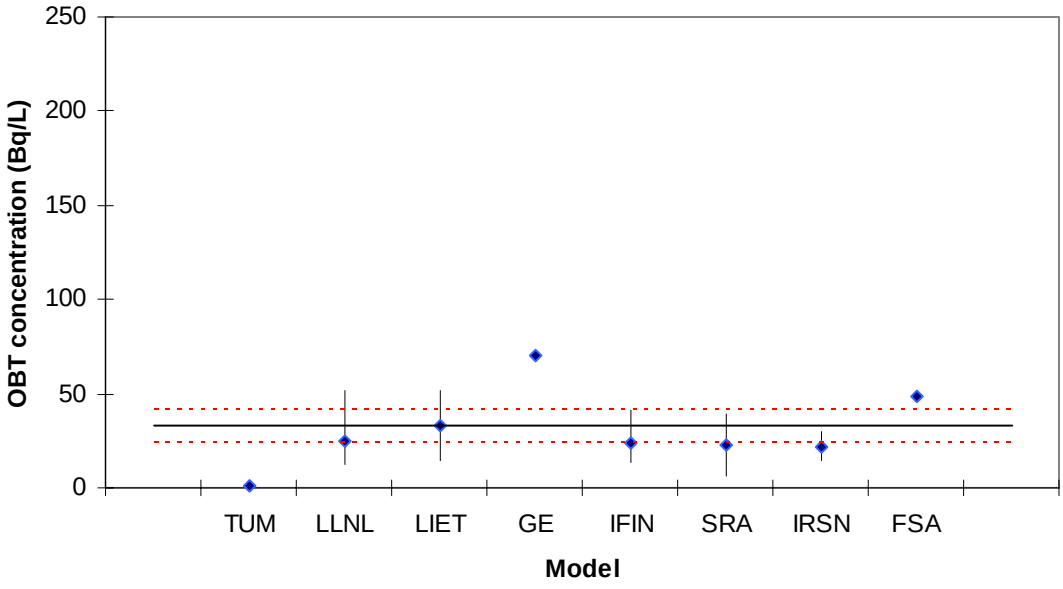

Figure A2b. As in Fig. A2a but predictions were obtained using air concentrations calculated from monthly meteorological data (daylight hours only)

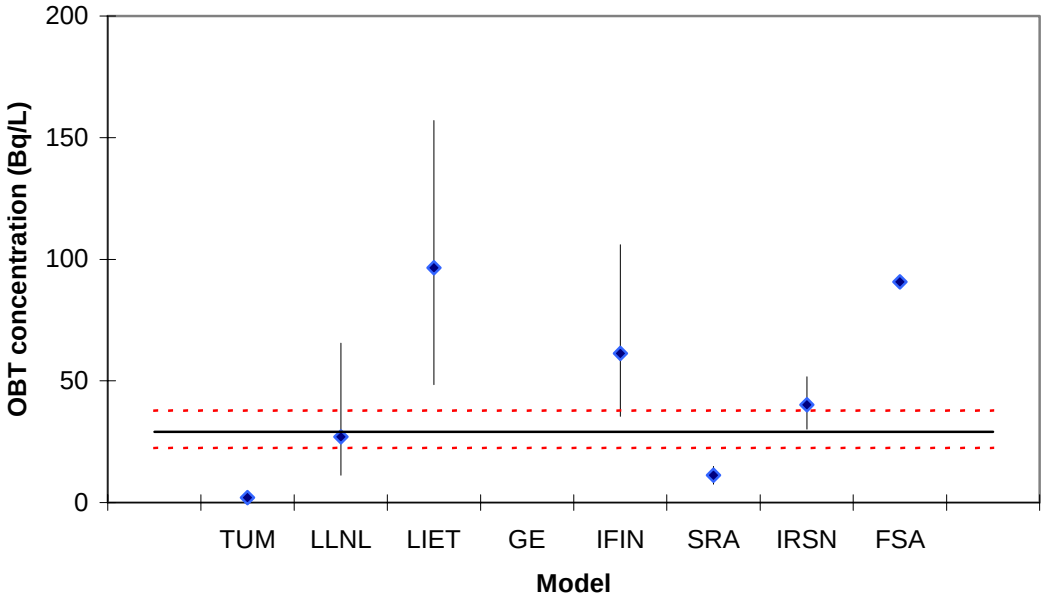

Figure A3a. Average OBT concentrations in calf flesh and heart at DF8 in September, predicted using the air concentrations given in the scenario description.

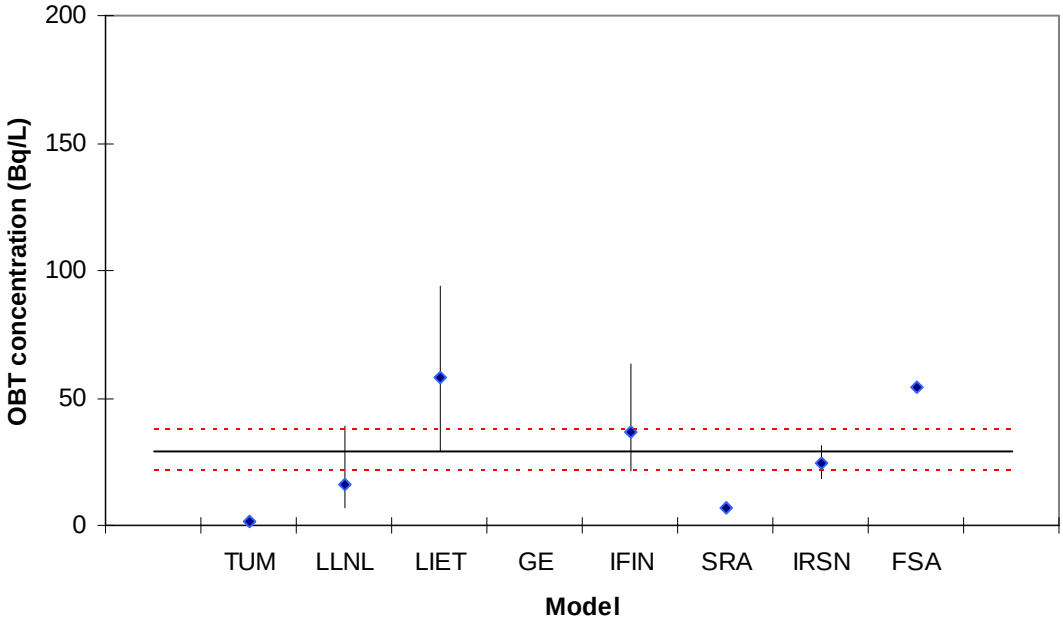

Figure A3b. As in Fig. A3a but predictions were obtained using air concentrations calculated from monthly meteorological data (daylight hours only)

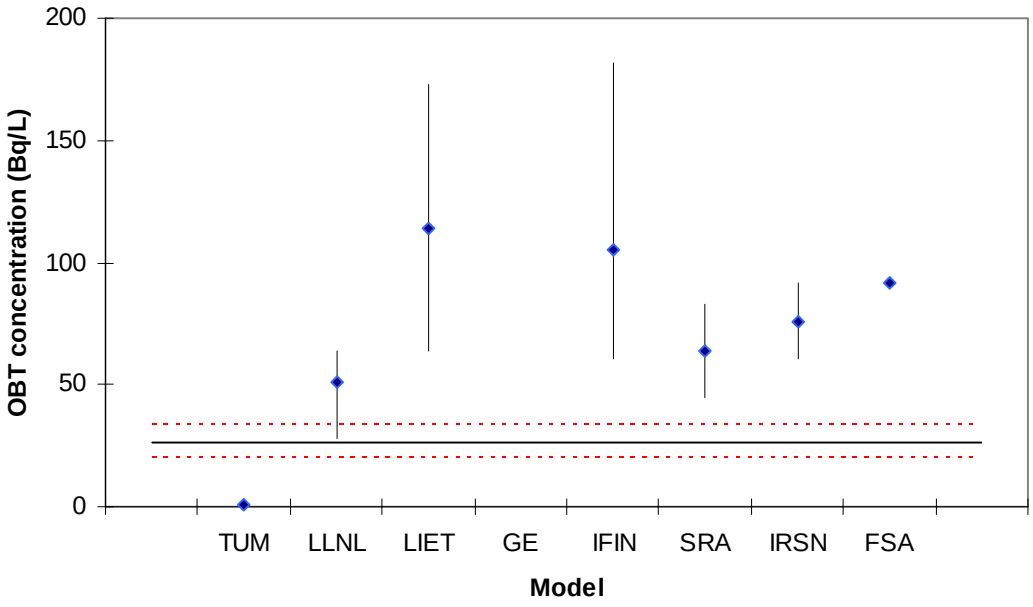

Figure A4a. Average OBT concentration in eggs at F27 in September, predicted using the air concentrations given in the scenario description.

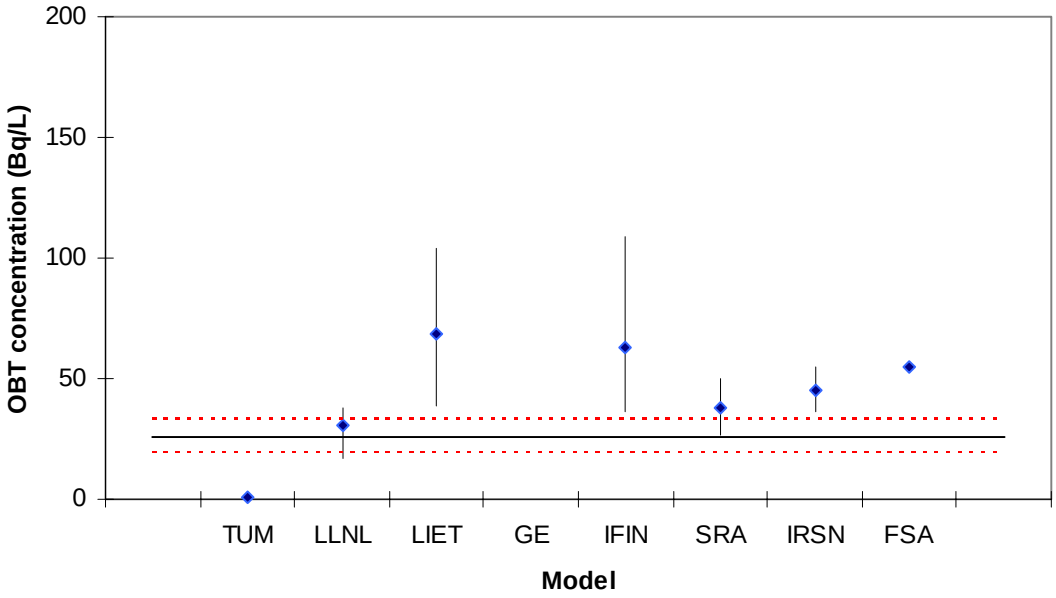

Figure A4b. As in Fig. A4a but predicted using air concentrations calculated from monthly meteorological data (daylight hours only)

Not all models produced better results when they were driven by daylight air concentrations. A model developed by AECL, which was run specifically to investigate the effects of averaging time on model predictions, achieved more accurate results using the 24-hour concentrations (Table A2). The AECL model is similar to the LLNL model but is designed to be realistic rather than conservative. It produces lower concentrations for most scenario endpoints than the models in the study, and the use of daylight air concentrations resulted in predictions that were lower than the observations by a factor of 2. Thus conclusions regarding the best averaging time for the air concentrations appear to be model dependent and more work is required to determine whether the 24-hour or daylight averaging period is most appropriate. This question is directly related to the amount of OBT that is formed at night. If most OBT is produced during the day, the models should be run with daylight air concentrations. If significant amounts of OBT are produced at night, the 24-hour concentrations would be more appropriate.

Table A2. Predicted to observed ratios using the AECL model averaged over all sampling sites and sampling times

Endpoint	Averaging Time	
	24 hours	Daylight hours
Plant OBT	1.18	0.40
Animal HTO	0.95	0.32
Animal OBT	1.10	0.37

APPENDIX B

Pickering Scenario Description – Revision 1
EMRAS Tritium/C14 Working Group

2004 June

BACKGROUND

Small amounts of tritium are released continuously from the CANDU reactors that make up Pickering Nuclear Generating Station (PNGS) on the north shore of Lake Ontario. The releases have been going on for many years and concentrations in various parts of the environment are likely to be in equilibrium. A large number of environmental and biological samples were collected in 2002 from four sites in the vicinity of the station. HTO concentrations were measured in air, precipitation, soil, drinking water, plants (including the crops that make up the diet of the local farm animals) and products derived from the animals themselves; OBT concentrations were measured in the plant and animal samples. These data are offered here as a test of models that predict the long-term average tritium concentrations in terrestrial systems due to chronic releases.

SITE DESCRIPTION

PNGS is made up of two units, each consisting of four reactors. Unit A has been shut down for several years but still releases significant amounts of tritium. Unit B was running at full power during the study period. The land surrounding the station is gently rolling and supports a mixture of uses, including industrial, recreational, agricultural and residential.

The samples were taken at two dairy farms (DF8 and DF11), a hobby farm (F27) and a small garden plot (P2) (Figure 1 and Table 1). All of the sampling sites were located to the northeast of PNGS; the two dairy farms lay about 10 km from the station, the hobby farm about 7 km and the garden plot about 1 km. As dairy farms, DF8 and DF11 yielded much the same sort of samples, including corn, pasture grasses, a variety of grains, milk and meat. In contrast, F27 produced mainly fruit, garden vegetables, chickens and eggs. A limited number of plants are grown at P2 for research purposes and raspberry leaves and grass were sampled.

Meteorological data for the Pickering area are given in Table 2. The air temperatures were measured locally in 2002. The solar radiation data represent long-term average conditions at Toronto, about 25 km west of Pickering. The precipitation data are long-term averages for the Pickering area. The fraction of time that rain falls when the wind blows toward F27 is 0.125; the analogous number for DF8, DF11 and P2 is 0.115. These frequencies are based on long-term average data for Toronto and are believed to be

overestimates. The average absolute humidity for the 2002 growing season for the area was 0.012 kg m^{-3}.

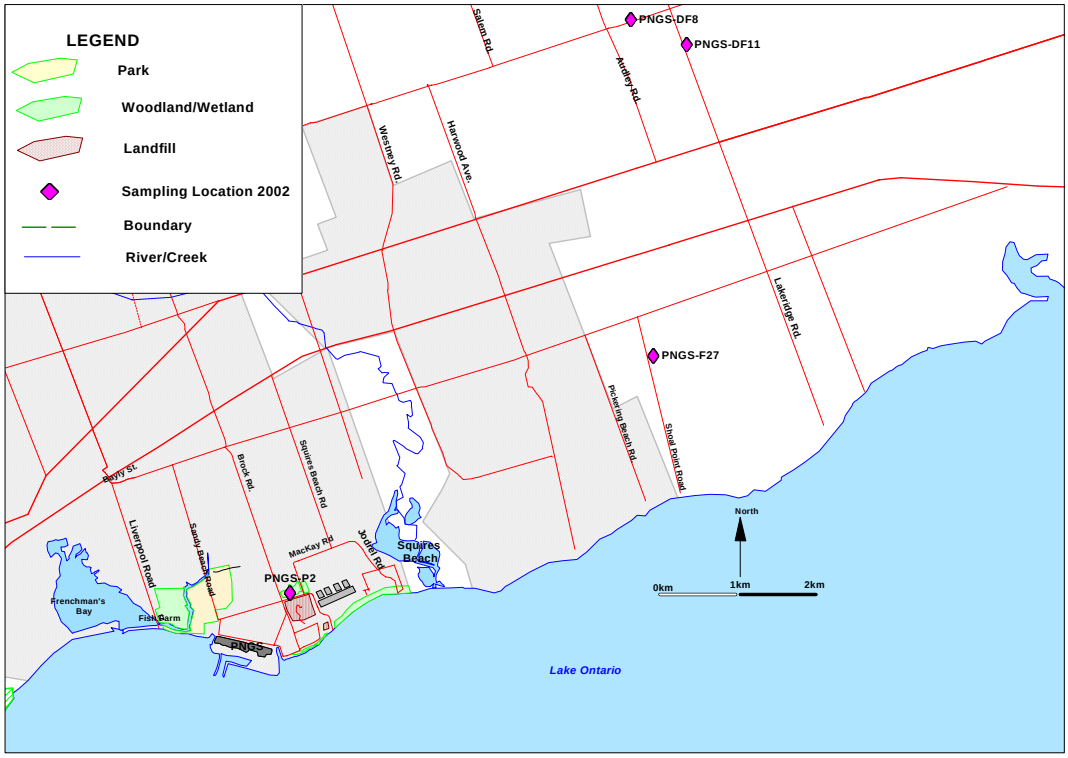

Fig 1. Map of the study area showing the tritium release points (red polygons) and sampling sites (green polygons).

Table 1. Location and description of the sampling sites

Site	Distance from Unit A	Description
DF8	10,520	Dairy farm, growing pasture grasses, corn and a variety of grains, and raising dairy cows
DF11	10,405	Dairy farm, growing pasture grasses, corn and a variety of grains, and raising dairy cows
F27	7,125	Hobby farm, growing fruit, pasture grasses and garden vegetables, and raising chickens
P2	1,150	Research garden plot growing berries and surrounded by grass

Table 2. Meteorological data for the Pickering area

Month	Air Temperature (C)		Solar Radiation (W m^{-2})		Rainfall
	Daily mean	Mean daily max	Daily mean	Mean daily max	(mm)
May	9.2	14.5	230	658	72.5
June	16.3	21.9	254	708	64.5
July	20.9	27.6	254	717	68.4
August	20.5	27.3	216	642	77.6
Sept	18.6	25.2	163	528	66.9

FARM PRACTICES

The cows at DF8 and DF11 are fed total mixed ration (TMR), a blend of various feeds harvested in the previous year. The make-up of the TMR at the two farms is shown in Table 3. The corn silage, feed corn, baled hay, haylage and barley are all obtained locally. The silos containing corn silage are filled annually in September. The haylage silos are filled two to three times per year, depending on the growing season. All of the other feed components (brewer's grain, dairy supplement, limestone) are purchased from remote locations and are assumed to contain only background levels of tritium. The total food intake by the cows was estimated by the owners to be 19.0 and 8.8 kg dry weight per day for farms DF8 and DF11, respectively. The latter value is believed to underestimate the true intake.

Table 3. Ratios of feed components in TMR

Type of feed	DF8 (%)	DF11 (%)
Corn silage	45.5	41.9
Feed corn	13.9	22.9
Haylage	12.7	19.6
Brewer's grain	12.7	0
Dairy supplement	7.4	13.8
Baled (dried) hay	4.6	1.9
Barley	3.0	0
Limestone	0.1	0

The chickens raised at F27 were essentially free-range and their food intake was not regulated or monitored. As a result, the make-up of their diet and their intakes could only be estimated (Table 4). The feed corn in their diet was purchased from DF11, and the "other sources" consisted largely of table scraps.

Table 4. Estimated composition of the chicken diet at F27

Type of Feed	% of Diet
Grass	10
Chicken greens (leafy material such as lettuce, beet tops, etc.)	10
Feed corn	30
Oyster shells	3
Apples	5
Carrots	5
Potatoes	5
Green beans	7
Other sources	25

The amount of drinking water ingested by the cows and chickens was not monitored. Irrigation was not carried out to any significant extent at any of the farms during the study period.

TRITIUM MEASUREMENTS

All of the samples were collected in two field campaigns carried out in 2002, the first from July 8 to 10 and the second from September 16 to 18. All of the samples collected in July were oven-dried before the HTO could be extracted and so were suitable for OBT

analysis only. The September samples were frozen in their fresh state and were analysed for both HTO and OBT.

Air: Air concentrations at the sites are measured routinely as part of a monitoring program carried out by the utility. Active molecular sieve samplers provided monthly-average concentrations at P2 and annual average concentrations were available from passive diffusion samplers at the other sites. The background air concentration due to tritium sources other than PNGS is 0.19 Bq m^{-3}. Tritium concentrations in the samples were determined using liquid scintillation counting (LSC) techniques.

Precipitation: Precipitation is collected monthly by the utility at DF8, F27 and P2 using gauges with an oil layer to prevent the transfer of tritium between air and water. The water collected was analysed for its tritium content using LSC.

Plants: At the farm sites, samples were collected of each of the plants that made up the animal diets, as well as separate samples of TMR. At F27, additional measurements were made of garden vegetables, root crops and fruit. Table 5 lists the samples collected and their measured water contents. Water equivalent factors (the fraction by weight of water produced when a dry sample is combusted) are also listed. However, these are literature values since the measured values seem low, likely because of the difficulty in collecting all of the water following combustion. Published values of plant yields are also shown in Table 5 for those crops for which data are available. The water in the September samples was extracted by freeze-drying, and HTO concentrations were determined by LSC. The dry matter in the July and September samples was washed with tritium-free water and then oven-dried and combusted in a combustion bomb. LSC of the combustion water yielded non-exchangeable OBT concentrations.

Animal Products: The meat samples from DF8 and DF11 came from calves that were either stillborn or died from complications at birth. The mothers were three years old or younger and were raised exclusively on these farms. A local veterinarian dissected the calves and provided samples of flesh and heart. Additionally, composite milk samples consisting of a mixture of milk from all cows in the herd were collected in July at both farms.

The only animal products sampled at F27 in the July campaign were eggs. Two eggs from mature layers (24-65 weeks old) were combined and a further measurement was made of a composite sample of about 12 eggs. In addition, an immature egg taken from the body cavity of a slaughtered chicken was analysed. In September, in addition to eggs, blood and flesh were also analysed from a single chicken that was probably less than 24 weeks old, as there were no mature yolks in its body cavity. HTO and OBT concentrations in all animal products were determined using the same procedures as for plants.

Table 5. Measured water contents and published yields and water equivalent factors for the sampled crops

Crop type	Site	Month	Plant type	Water content (%)	Water equivalent factor	Yield (kg fw m^{-2})
Forage	DF8	Jul	Hay[¤]	78.4	0.587	0.47
			Haylage[¤]	70.5	0.594	0.47[§]
			Barley	10.5	0.567	0.28
			TMR*	51.9	0.582	
		Sep	Alfalfa	76.4	0.592	0.40
			Baled hay[¤]	13.8	0.584	0.47[§]
			Corn silage	61.5	0.579	2.7[§]
			Haylage	63.7	0.594	0.47[§]
			Feed corn	25.2	0.572	2.7
			Barley	12.6	0.567	0.28
			Soya meal	11.6	0.600	0.24[§]
			TMR	54.9	0.582	
	DF11	Jul	Alfalfa	78.0	0.592	0.40
			Baled hay	15.9	0.584	0.47[§]
			Haylage	34.5	0.594	0.47[§]
			Feed corn	20.1	0.572	2.7
			TMR*	41.7	0.578	
		Sep	Alfalfa	73.0	0.592	0.40
			Baled hay	11.5	0.584	0.47[§]
			Corn silage	60.2	0.579	2.7[§]
			Haylage	36.9	0.594	0.47[§]
			Feed corn	22.4	0.572	2.7
			TMR*	39.2	0.578	
	F27	Jul	Grass	56.1	0.587	
			Spring wheat	13.3	0.617	0.33
			Soya meal	10.8	0.600	0.24[§]
		Sep	Grass	76.1	0.587	
			Feed corn	5.0	0.572	2.7
			Spring wheat	10.0	0.617	0.33
			Soya meal	6.0	0.600	0.24[§]
	P2	Sep	Raspberry leaves	54.8	0.470	
			Grass	75.9	0.587	
Garden vegetables	F27	Jul	Mixed vegetables[‡]	87.4	0.537	
		Sep	Tomato	81.0	0.543	2.0
			Cucumber	94.0	0.520	1.7
Fruit	F27	Sep	Apple	80.0	0.575	1.9
			Pear	83.2	0.560	0.68
			Raspberry	85.1	0.562	0.16
Root crops	F27	Sep	Carrots and potatoes	81.1	0.543	3.0
			Beet	87.4	0.523	2.3
			Garlic	55.3	0.549	1.7

[¤] hay refers to fresh cut pasture; baled hay is dried pasture; haylage is hay that has been stored in a silo

* produced in 2001

[‡] beet, cabbage, hot pepper, onion, dill, potato, spinach

[§] yield of parent plant in the field

The animal products sampled during the study are listed in Table 6, together with measured water contents and literature values of the water equivalent factors.

Table 6. Measured water contents and published water equivalent factors for the sampled animal products

Site	Month	Animal product	Water content (%)	Water equivalent factor
DF8	Jul	Milk	85.9	0.746
	Sep	Calf flesh	75.7	0.646
		Calf heart	76.6	0.753
DF11	Jul	Milk	87.5	0.746
	Sep	Calf flesh	75.5	0.646
		Calf heart	76.3	0.753
F27	Jul	Egg	74.8	0.803
		Composite egg	71.5	0.803
		Immature egg	47.2	0.803
	Sep	Egg	76.0	0.803
		Chicken blood	80.0	Unknown
		Chicken flesh	74.4	0.697

Drinking Water: Samples of water were taken from the deep wells that supply drinking water for the cows at farms DF8 and DF11 in the September sampling period. Concentrations in drinking water at F27, which comes from a shallow well, are available from routine monitoring by the utility, but not for each month. The value given below in Table 8 is the average for June to December.

Soil: Soil cores were collected at a single location at each site. Three cores 15 cm in diameter and 5 cm deep were taken at each location and composited for analysis. The cores were collected from undisturbed locations in grassed areas or where the soil had lain fallow for some time. No detailed analysis of physical properties was done but the soils at DF8, DF11 and P2 are believed to be loams or clay loams with bulk density, pH and organic content around 1.08 g cm^{-3}, 7.3 and 5.2% dry weight, respectively. At F27, where the cores were taken beside a road, the soil contained more sand. The samples were analysed for their HTO and OBT concentrations using the procedures discussed above for plant and animal samples. Water contents are listed in Table 7.

Table 7. Water content of the sampled soils (% wet weight)

	DF8	DF11	F27	P2
July	-	12.9	25.9	-
September	19.4	14.0	15.0	26.1

Uncertainties: The observed concentrations in all environmental compartments were relatively low, although they were at least a factor 4-5 above background. Counting errors for both HTO and OBT samples were less than 10% in most cases. A further error of perhaps 30% must be added to the air concentrations to account for the uncertainty in the passive diffusion sampler data at DF8, DF11 and F27. An additional uncertainty of about 30% must also be added to the plant and animal concentrations to account for natural variability.

INPUT DATA

Best estimates of the HTO concentrations in air and drinking water at the study sites are shown in Table 8. HTO concentrations in monthly precipitation are given in Table 9.

Table 8. Measured HTO concentrations in air and drinking water. The air concentrations include a background contribution of 0.19 Bq m^{-3}.

Compartment	DF8	DF11	F27	P2
Air concentration (Bq m^{-3})				
2002 May	1.01	1.01	1.56	24
June	1.39	1.39	2.14	33
July	0.93	0.93	1.43	22
August	0.88	0.88	1.36	21
September	0.67	0.67	1.04	16
Air concentration (Bq m^{-3})				
2001 May	0.49	0.49	0.77	12
June	2.83	2.83	4.40	69
July	0.86	0.86	1.34	21
August	1.23	1.23	1.92	30
September	0.66	0.66	1.02	16
Drinking water concentration (Bq L^{-1}) 2002 September	18.6	21.1	24.3*	Not relevant

* average value for June-December 2002

Table 9. Measured monthly HTO concentrations in precipitation

Month	HTO Concentration in Precipitation (Bq L^{-1})		
	DF8	F27	P2
January	not available	not available	3670
February	not available	18	1350
March	not available	24	347
April	24	29	474
May	69	14	525
June	85	61	579
July	9	14	205
August	49	19	442
September	13	22	452

SCENARIO CALCULATIONS

From the information provided above, calculate

(i) HTO and non-exchangeable OBT concentrations in the plants and animal products listed in Tables 5 and 6. 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 (Bq L^{-1}) concentrations in the top 5-cm soil layer for each site for each sampling period.

(iii) 95% confidence intervals on all predictions.

The predicted HTO concentrations in plants should reflect average conditions over the growing season and not the measured concentrations at the sampling times. HTO is very mobile in plants and concentrations are strongly dependent on the air concentration in effect in the few hours before sampling. Since these concentrations (or the meteorological and source term data required to calculate them) are not available, no attempt will be made to compare predicted and observed HTO concentrations in plants. Rather, the predictions will be used to help explain differences among model results for OBT concentrations.

APPENDIX C

MODEL DESCRIPTIONS

LLNL Model

Introduction

The stochastic model DCART (Doses from Chronic Atmospheric Releases of Tritium) was used to generate predictions for the Pickering scenario. DCART was developed as a realistic assessment model to be used in a dose reconstruction for tritium releases from the Lawrence Livermore National Laboratory. It is a steady-state, analytical compartment model that calculates uncertainties using parameter distributions and Latin Hypercube Sampling. Compartments include air and air moisture, soil, plant water, plant organic matter, animal water, and animal organic matter.

In the plant, processes include uptake of tritiated water (HTO) from soil water and air moisture and conversion to organically bound tritium (OBT); in the animal, processes include inhalation and skin absorption of air moisture, ingestion of water and food, and the partitioning into HTO and OBT within the animal. For the Pickering Scenario, starting with concentrations of HTO in air (Bq m^{-3}), concentrations of HTO and OBT in vegetables (leafy, fruit and root), fruit, pasture, grain, cow milk, beef, chickens and eggs were calculated; HTO concentrations in soil were also calculated.

Key assumptions (unique to the Pickering Scenario)

Four sets of calculations had to be carried out in order to predict the list of items requested in the scenario description. Calculations using air concentrations from 2001 were made to estimate concentrations in the components (e.g., barley, corn, haylage) of the total mixed ration (TMR). Calculations using air concentrations from 2002 were made to estimate the concentrations in various types of fodder, vegetables and fruit. Calculations using air concentrations from 2002 and TMR from 2001 were made to estimate concentrations in milk and calf meat. If the product calculated in 2002 was harvested in July, the mean of the air concentrations from May to July was used; if the product calculated in 2002 was harvested in September, the mean of the air concentrations from May to September was used.

Modeling Approaches

DCART would normally be used to estimate annual mean concentrations in plant and animal products from annual mean air concentrations. Shorter periods of time, such as those for the Pickering scenario, may also be modeled, as long as the averaging time is long enough for the system to approach equilibrium.

DCART is calibrated so that the ratio of soil moisture concentration to air moisture concentration is a set fraction for a release of HTO. Alternatively, soil water concentrations may be set equal to concentrations in rainfall. DCART was run with both

assumptions for the September 2002 predictions and yielded almost identical results in plants and animals in both cases. This is not surprising given that soil water is not an important pathway in DCART and that soil water concentrations differed by less than a factor of two.

The mean concentrations of HTO in plant water of leafy vegetables and pasture are given by the equation:

$$C_{pw} = 1/\gamma \ [R_H \, C_{a\_HTO} / H_a + (1 - R_H) \, C_{sw}]$$

where:

C_{pw} = concentration of tritium in the plant water (Bq L^{-1} or Bq kg^{-1})
γ = ratio of vapor pressure between HTO and H_2O (0.909)
R_H = relative humidity
C_{a_HTO} = concentration of HTO in air (Bq m^{-3})
H_a = absolute humidity (kg m^{-3})
C_{sw} = concentration of tritium in soil moisture (Bq L^{-1})

The mean concentrations of HTO in fruits, fruit vegetables and grain are calculated similarly but without accounting for γ or relative humidity. Instead it is assumed that 60% of the HTO in the fruit comes from air moisture and the other 40% comes from soil water. The annual concentration of HTO in a root crop is assumed to equal 95% of the concentration in soil water.

Mixed vegetables were assumed to be equal proportions of beets, cabbage, hot pepper, dill, spinach, onion, and potato.

Concentration of OBT in all plants in Bq L^{-1} water equivalent was assumed to equal the concentration in plant water (as calculated for leafy vegetables and pasture) reduced by a discrimination factor that arises from isotopic effects in OBT formation. Because of this assumption, in DCART all concentrations of OBT in all types of vegetables are the same in Bq L^{-1}, given the same air concentration.

In DCART it has been assumed that the HTO concentration in the animal (in Bq L^{-1}) equals the weighted concentration of tritium obtained from food, water and air (all also in Bq L^{-1}) taken in by the animal. In other words, DCART calculates the Bq L^{-1} HTO in the animal from the concentrations in and fractions of water contributed by plant water, plant organic matter, drinking water, and inhalation and skin-absorption. OBT concentrations in the animal are assumed equal to HTO concentrations.

DCART calculates tritium concentrations in plants and animals in both Bq kg^{-1} fresh weight and Bq L^{-1} water or water equivalent. To convert between the two requires parameters for dry matter content and water equivalent.

Air concentration in Bq m^{-3}, obtained either from measurements at the location of interest or from dispersion modeling, is the primary input to DCART. A value for absolute

humidity is needed to convert tritium in air volume to tritium in air moisture, which is the parameter that drives all calculations. All plants and animals are assumed exposed to the same air concentration, regardless of whether this is physically possible.

Parameter values used in the Pickering Scenario

Air: The mean air concentrations and standard deviations in Bq m^{-3} that were used for different time periods are shown below. Distributions are lognormal.

	Air concentrations in Bq m^{-3}			
	DF8	DF11	F27	P2
2001	1.21 ± 0.944	1.21 ± 0.944		
2002 May-July	1.15 ± 0.344	1.15 ± 0.344	1.77 ± 0.53	
2002 May-Sept.	0.976 ± 0.263	0.976 ± 0.263	1.51 ± 0.403	23.2 ± 6.22

The absolute humidity used for Pickering was 0.012 kg m^{-3} with an uncertainty on a normal distribution of ± 10%. The relative humidity used for Pickering was 73% with a normal distribution ± 10% uncertainty.

Soil:

- A triangular distribution has been applied to the calibrated fraction that relates soil water concentration to concentration in air moisture; the minimum is 0.1, the best estimate 0.3, and the maximum 0.5.
- For 2001 calculations, soil was only assumed to have one-third the concentration of air moisture. For 2002, predicted soil concentrations based on 30% air moisture are compared below to soil concentrations based on rainfall concentrations. Concentrations are given in Bq L^{-1}, and distributions are lognormal.

	Soil water concentrations in Bq L^{-1}	
	Based on 0.3 x air moisture	Based on rainfall
DF8 and DF11	24.5 ± 9.70	41.5 ± 31.2
F27	37.8 ± 16.2	25.1 ± 15.3
P2	580 ± 238	432 ± 123

Plant:

- An isotopic discrimination factor for OBT of 0.7 has been chosen. This parameter has an extreme value distribution with a 2.5% confidence limit of 0.49 and a 97.5% confidence limit of 1.18 based on empirical data.
- The fractional relationship between concentration of HTO in fruit and grain and concentration in air moisture is described by a triangular distribution (0.5 – 0.6 – 0.7).

Animals:

- For the calculation of OBT alone, because the transfer of plant OBT to animal OBT occurs preferentially, at least dynamically, the part of the equation that accounts for this transfer has been multiplied by a parameter with value 1 ± 40%; the lower bound is truncated at 0.8. This parameter obviously does nothing to change the best estimate; it only increases the uncertainty about the concentrations of OBT.
- A parameter was added to DCART to help account for the natural variability between an average cow and the single individuals sampled for the scenario at farms DF8 and DF11. This parameter has a normal distribution with a value of 1 and an uncertainty of ± 30%.
- Ingestion and inhalation parameters are shown in the table immediately below. Uncertainties have normal distributions unless noted.

Diet (kg dw d^{-1})	Cows – DF8	Cows – DF11	Chickens**/eggs
TMR*	19 ± 0.95	16.4 ± 1.64	
Green beans			0.0135
Feed corn			0.0580
Grass etc.			0.0386
Apples			0.00966
Carrots and potatoes			0.01932
Water	80 ± 8		0.29
Inhalation (m^3 d^{-1})	144 ± 67; left truncated at 74		1 ± 0.6 truncated at 0.3 and 2.0

* TMR is made up of varying amounts of baled hay, corn silage, feed corn, haylage and, for D8 only, barley.

** The uncertainty on the diets for chickens is rectangular with the limits being ± 25% of the best estimate; water ingestion for chickens has a triangular distribution with minimum at 0.15 and maximum at 0.44.

- To estimate the concentration in TMR, the concentration of the food types making up TMR were calculated for 2001 air concentrations on a Bq kg^{-1} basis for both HTO and OBT. Total HTO or OBT in the daily diet of TMR was calculated by summing the products of the concentration of each foodstuff times the ingestion rate-equivalent for that foodstuff in TMR. Then the Bq kg^{-1} TMR HTO or OBT was calculated by dividing total Bq d^{-1} HTO or OBT by kg TMR d^{-1}. Concentration of TMR in Bq kg^{-1} was then converted to Bq L^{-1}. The TMR from 2001 was input into DCART as animal feed in 2002. The uncertainty on the concentration of TMR was lognormal ± 45%.
- The uncertainty on the drinking water concentrations was ± 20% for a normal distribution.

Application of the model to the scenario

Air concentrations (Bq m^{-3}) used as input were obtained from the scenario description. The monthly values were averaged, and the standard deviations of the averages were used to estimate uncertainty; this means that the uncertainty about the mean is over-estimated.

Concentrations for May to July were weighted averages (four weeks in May, four weeks in June, and two weeks in July).

The average absolute humidity was taken from the scenario description. Relative humidity was calculated partly using temperatures from Table 2. Rainfall concentrations from Table 9 were averaged to estimate possible concentrations in soil water.

Animal diets were based on the information provided in the scenario description. Using the ratios of each type of feed in TMR (from Table 3), a diet of TMR was devised that added up to 19 kg dry weight for Farm DF8 and 16.4 kg dry weight for DF11 (revised upwards from the acknowledged low limit of 8.8 kg). The revised diet for DF11 is reasonable, although it is still less than the diet at DF8: metabolizable calories were estimated at 39 Mcal for DF8 and 20 Mcal for DF11. Concentrations for only the feed grown locally (baled hay, barley, corn silage, feed corn and haylage) were calculated. It was assumed that at each dairy farm the milk cow and the calf ate the same quantity and composition of food, because the concentration in the adult or juvenile would have been nearly the same had the diet for the calf been proportionally smaller; at F27, the chicken and the laying hen ate the same diet. The diet of the chickens was also estimated from Table 4 in the scenario description and the assumption that chickens ate 0.139 kg per day. Drinking water concentrations were taken from Table 8 in the scenario description.

For this scenario, the values for dry matter and water equivalent given in Tables 5 and 6 of the scenario description were averaged between themselves, when appropriate, and with other values from a database. After combining the various values reported, small uncertainties were applied.

Sensitivity

Sensitivity analyses were run for various endpoints. In DCART, all types of fodder were modeled as just two groupings based on whether they were derived from foliage or grain:

Group 1. Alfalfa hay, haylage, baled hay and corn silage
Group 2. Soya meal, barley and feed corn

The sensitivity of these two categories to various parameters is essentially identical. Parameters having rank correlation coefficients greater than 0.2 are shown below for the categories of fodder.

HTO in			OBT in	
Group 1	Group 2	Parameter	Group 1	Group 2
0.91	0.92	Air concentration (Bq m^{-3})	0.73	0.73
NA	NA	Isotopic discrimination	0.56	0.56
-0.29	-0.30	Absolute humidity	-0.24	-0.24

When dairy and meat cows are fed a diet of TMR, the important parameters and their rank correlation coefficients greater than 0.2 from a sensitivity analysis are:

HTO in			OBT in	
Milk	Meat	Parameter	Milk	Meat
0.86	0.87	Natural variability	0.72	0.73
NA	0.51	Plant OBT to animal OBT	NA	0.51
0.31	0.32	Drinking water concentration	0.26	0.26
0.28	0.26	Concentration of HTO in TMR	0.23	0.22

The two parameters to which the endpoints are most sensitive (above) are those that attempt to account for uncertainty that cannot be quantified easily.

For the various vegetables eaten by the chickens, the sensitive parameters were similar to those above except that, for root crops, the soil concentration parameter had a rank correlation coefficient of 0.58, and the parameter relating concentration in potato to concentration in air moisture had one of 0.22. The parameter for relative humidity had a rank correlation coefficient of 0.22 for grass.

The uncertainty about natural variability was neglected for chickens and eggs because there is much less variability in chickens than in cows. The rank correlation coefficients for chicken and eggs are as similar as those for milk and meat, so only those for chicken are shown below.

Chicken HTO	Parameter	Chicken OBT
0.75	Air concentration (Bq m^{-3})	0.49
NA	Plant OBT to animal OBT	0.73
0.39	Drinking water (Bq L^{-1})	0.26
-0.29	Drinking water rate (L d^{-1})	-0.20
-0.22	Absolute humidity	-0.15

Reference

Peterson, S-R. 2004. Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL) Part 1. Description of Tritium Dose Model (DCART) for Chronic Releases from LLNL. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-205083. The report is available at http://library.llnl.gov/uhtbin/cgisirsi/0/0/60/55/X.

IFIN Model

1. Introduction

Model name: IFIN_Pick
Purpose of the model: IFIN_Pick is an assessment model. It was designed to not underpredict and to overpredict by no more than a factor of 3.
Type of model: simple, steady-state, analytical.
Biological/environmental compartments considered: air, precipitation, soil water, plants and animal products.
Transport processes – HTO transfer from air to precipitation, soil, plants and animal products. OBT formation in plants and transfer to animals. Simplified transfer coefficients are used throughout.
Endpoints: HTO concentrations in soil, plants and animal products; OBT concentrations in plants and animal products, as required by the scenario.

2. Model Formulation and Key Assumptions

The HTO concentration in soil water is equal to the concentration in rain (60% from the current month and 40% from the previous month) plus 10% of the concentration in air moisture.

The HTO concentration in plant water (C_{pw}) is given by the classic formula (Murphy 1984)

$$C_{pw} = 1.1\ [RH\ C_a + (1\text{-}RH)\ C_s]$$

where RH is the relative humidity, C_a is the water vapour HTO concentration and C_s is the soil water HTO concentration.

For forage crops, leafy vegetables and TMR, the OBT concentration in the plant combustion water equals the HTO concentration in the plant water averaged over the previous 3 months. The OBT/HTO ratio for fruit and root crops is 0.75 and 0.40 respectively.

86% of TMR is made up of contaminated feed, with 70% coming from the July harvest and 30% from the September harvest.

Animals take in HTO with their drinking water and their food. OBT intake occurs only with food. The cow at DF11 was assumed to have the same diet as the cow at DF8.

HTO concentrations in milk, eggs and meat (C_{an}) were calculated from

$$C_{an}^{HTO} = \text{FHH*HTO\_intake} + \text{FOH*OBT\_intake},$$

where HTO_intake and OBT_intake are the intake rates of HTO and OBT respectively, and FHH and FOH are animal-specific transfer factors derived from a metabolic model (see table below). Similarly, OBT concentrations in animal products are calculated from

$$C_{an}^{OBT} = \text{FHO*HTO\_intake} + \text{FOO*OBT\_intake} .$$

	FHH	FOH	FHO	FOO
Cow milk	0.01	0.007	0.0003	0.007
Veal meat	0.028	0.02	0.002	0.05
Broiler meat	2.6	2.3	0.2	3.1
Egg	2	1.7	0.13	2.4

To convert OBT concentrations in fresh weight to concentrations in combustion water, it was assumed that the dry fraction was 0.13 for milk, 0.28 for veal meat and 0.26 for hens and eggs. The water equivalent was 0.7 for milk, 0.66 for veal meat and 0.8 for egg and hens.

3. Uncertainties

Uncertainties in the model predictions were estimated by expert judgement and are believed to be optimistic.

LIETDOS Model

Tritium Concentrations in Air and Soil

LIETDOS is a compartment model for a terrestrial system that has achieved steady state in terms of activity exchange by balancing gains and losses. Taking into account that HTO is present mainly in the aqueous phase of a compartment, the total compartment inventory and the water-phase inventory of tritium are assumed to be the same in soil and in air.

The simultaneous balancing of gains and losses for both soil and air compartments allows the activity inventories of the two compartments to be calculated as follows:

$$S + \lambda_{sa}\, Q_{soil} - \lambda_{air}\, Q_{air} = 0; \quad (1)$$

$$\Lambda_{as}\, Q_{air} - \lambda_{soil}\, Q_{soil} = 0, \quad (2)$$

where S represents the rate of HTO input (i.e., the HTO emission rate) into the air compartment (Bq/d); Q_{soil} and Q_{air} represent the compartment HTO inventory in soil and air respectively (Bq); λ_{sa} is the soil to air activity transfer rate constant (d^{-1}); Λ_{as} is the air to soil activity transfer rate constant (d^{-1}); and λ_{air} and λ_{soil} are the effective activity decrease rate constants in air and soil respectively (d^{-1}).

Using equation (2), the long-term average pollutant concentration in air (C_{air}, Bq m^{-3}) can be presented in the following manner:

$$\Lambda_{as}\, h_{air}\, C_{air} - \lambda_{soil}\, h_{soil}\, C_{soil} = 0, \quad (3)$$

where h_{air} is the atmospheric mixing height (m); h_{soil} is the soil compartment depth (m); and C_{soil} is the volumetric tritium activity in the soil (Bq/m^3 soil). λ_{soil} is the effective activity decrease rate constant in the soil compartment due to evapotranspiration ($\lambda_{evapotrans}$), recharging (λ_{sink}), runoff (λ_{runoff}) and radioactive decay (λ_r) (d^{-1}):

$$\lambda_{soil} = \lambda_{evapotrans} + \lambda_{sink} + \lambda_{runoff} + \lambda_r \quad (4)$$

Instead of using the air concentration C_{air}, the activity in precipitation (C_{pr}, Bq/m^3 water) has been used as input to the model. Based on this proposal, the activity balance equation for the soil compartment can be written as:

$$I_{pr} \cdot \phi_{soil} \cdot C_{pr} - \lambda_{soil} \cdot h_{soil} \cdot C_{soil} = 0; \quad (5)$$

where I_{pr} is the average precipitation rate during the time period of interest (m d^{-1}) and ϕ_{soil} is the volumetric soil moisture content (m^3 water per m^3 soil). According to equations (3) and (5)

$$\Lambda_{as} = I_{pr} \cdot \phi_{water} / (h_{air} \cdot \phi_{air}) \quad (6)$$

where ϕ_{water} is the activity scavenging factor for raindrops passing through air (m^3/m^3) and ϕ_{air} is the volumetric fraction of water in air (m^3/m^3). ϕ_{air} is given by

$$\phi_{air} = f_{RH} \cdot e_{sat} \cdot (M_{H2O}/\rho_{H2O}) / (R \cdot T_{air}) \quad (7)$$

where

$$e_{sat} = 100 \cdot \exp (11.28\text{-}2319.25/T_{air}) \quad (8)$$

$$f_{RH} = 100 \cdot e_{AH} / (M_{H2O} \cdot e_{sat} / (R \cdot T_{air})) \quad (9)$$

Here f_{RH} is the observed relative humidity (%); e_{sat} is the saturation vapor pressure (Pa); R is the universal gas constant = 8.31434 (Pa m^3)/(mol K); T_{air} is the ambient absolute air temperature (K); M_{H2O} is the molecular weight of water (18.016 g mol^{-1}); ρ_{H2O} is the density of water (10^6 g/m^3); and e_{AH} is the absolute humidity of the air (g m^{-3}).

The soil to air activity transfer rate constant (λ_{sa}), recharging coefficient (λ_{sink}) and runoff coefficient (λ_{runoff}) can be estimated according to known annual average evapo-transpiration, infiltration to ground water and runoff values $I_{evapotrans}$, $I_{recharge}$, I_{runoff} (m d^{-1}) respectively.

$$\lambda_{sa} = \frac{I_{evapotrans} \cdot \phi_{water}}{\phi_{soil} \cdot h_{soil}}; \quad (10)$$

$$\lambda_{recharge} = \frac{I_{recharge} \cdot \phi_{water}}{\phi_{soil} \cdot h_{soil}}; \quad (11)$$

$$\lambda_{runoff} = \frac{I_{runoff} \cdot \phi_{water}}{\phi_{soil} \cdot h_{soil}}. \quad (12)$$

The parameter values used in the calculations are presented in Table 1.

Table 1. Meteorological and soil properties for the PNGS environment.

Parameter	**Symbol**	**Mean value**
Air absolute humidity, g m^{-3}	e_{AH}	12
Mean atmospheric mixing height, m	h_{air}	600
Soil compartment depth, m	h_{soil}	2.5
Volumetric moisture content of the soil, L(water)/L(soil)	ϕ_{soil}	0.3
Annual average evapotranspiration, m d^{-1}	$I_{evapotrans}$	0.45
Annual average runoff, m d^{-1}	I_{runoff}	0.13

Using scenario data, the activity scavenging factor for raindrops passing through air (ϕ_{water}) was determined. The mean value during the time period 2001-2002 was 0.38.

HTO concentrations in the top soil layer for each site and each sampling period were calculated from Equations 1-12 and the parameter values in Table 1.

Tritium concentration in plant products

Equations are applied for leafy vegetable, pasture and hay according to IDRANAP (2002). The concentration of tritiated water in the leafy parts of plants ($C_{lv,w}$) is dependent on the tritium concentration in air moisture $C_{air,w}$ and soil water $C_{soil,w}$ according to

$$C_{lv,w}(HTO) = 1.1 \cdot f_{RH} \cdot C_{air,w}(HTO) + 1.17 \cdot (1 - f_{RH}) \cdot C_{soil,w}(HTO).$$

Other food items such as fruit vegetables, fruits, tubers and grain have a higher contribution by soil water and in those cases the HTO concentration was approximated by:

$$C_{other,w}(HTO) = 1.1 \cdot f_{RH} \cdot 0.33 \cdot C_{air,w}(HTO) + 1.17 \cdot (1 - f_{RH} \cdot 0.33) \cdot C_{soil,w}(HTO).$$

In our calculations, the concentration of OBT in combustion water, under equilibrium conditions, is related to the concentration of HTO in plant water by a factor 0.8:

$$C_{p,w}(OBT) = 0.8 \cdot C_{p,w}(HTO).$$

Tritium concentration in animal products

The concentration in animal products (C_{animal}, Bq kg^{-1}) depends on the transfer factor F (day kg^{-1}) and intake activity I (Bq/day):

$$C_{animal} = F \cdot I = F \cdot \Sigma u_i \cdot C_i$$

where u_i is the intake rate of diet item i (kg day^{-1}) and C_i is the concentration in that item (Bq kg^{-1}). In the case of tritium we considered two main chemical forms, HTO and OBT, including metabolic transformations between them. The concentration of HTO or OBT in animal products is

$$C_{HTO} = F_{HH} \cdot I_{HTO} + F_{OH} \cdot I_{OBT}$$
$$C_{OBT} = F_{HO} \cdot I_{HTO} + F_{OO} \cdot I_{OBT}$$

where F_{HH} is the transfer factor from HTO in food to HTO in animal product, F_{HO} is the transfer factor from HTO in food to OBT in animal product, F_{OH} is the transfer factor from OBT in food to HTO in animal product, and F_{OO} is the transfer factor from OBT in food to OBT in animal product.

Reference

IDRANAP. 2002. A standard guide for dose assessment of routine releases of tritium for any tritium facility. Center of Excellence Report WP3 IDRANAP 31-02/2002.

IRSN Model

Introduction

The IRSN Tocatta model simulates the transfer of tritium (and/or carbon-14) within terrestrial ecosystems in response to chronic or accidental releases of HTO (and/or carbon 14) to the atmosphere. It has been developed from bibliographical knowledge and in common with existing models of tritium transfer, in order to come within the conceptual and mathematical frameworks of the SYMBIOSE project[1].

Key Assumptions

- The model simulates the impacts of HTO releases to the atmosphere. HT and CH_3T releases are not considered;
- The main transfer paths of tritium within the terrestrial ecosystem are the following:
    - Net transfer of atmospheric HTO into the aqueous and organic parts of foliar systems (via diffusion/absorption and net photosynthesis, respectively);
    - Transfer onto soil via dry deposition and precipitation; HTO losses through evapotranspiration and vertical migration into the underlying soil layers (not considered here);
    - Transfer to animals by ingestion of vegetal products, inhalation and skin absorption, translocation and depuration (elimination).

Modeling Approaches

The conceptual modeling deals with splitting the continental biosphere into elementary components and identifying interactions (or transfer processes) between each component. This approach is based on the global interaction matrix shown in Figure 1.

[1] SYMBIOSE is a modelling and simulation platform for environmental pollutant risk assessment (IRSN, Cadarache, sponsored by Electricité de France).

	ATMOSPHERE	AGRICULTURAL ECOSYSTEM					
		SOIL	VEGETAL		ANIMAL Animal products		
SOURCE	Atmospheric release	Precipitation Dry deposition					
	AIR [HTO]		Absorption	Photosynthesis Translocation		Inhalation & Skin absorption	
		WAT [HTO]					Evaporation Transpiration Migration
			WAT [HTO]				
				O.M. [C14 & OBT]			
			VEGETAL		Ingestion	Ingestion Translocation	
					WAT [HTO]		
						O.M. [C14 & OBT]	
					ANIMAL		Elimination
							∞

Matrix of components (cross) and tranfer processes (vertical). WAT=water; O.M.=organic matter; ∞=all other systems, not considered here (e.g. underlying soil layers)

Figure 1: Conceptual model of the transfer of atmospheric HTO (and C-14) into the agricultural ecosystem.

In the case of accidental atmospheric releases, the mathematical modeling used for calculating daily inventories and fluxes is based on a system of first order differential equations expressing the conservation of tritium activity for each component:

$$\frac{d\left\{\chi_i [T]_i^{HTO}\right\}}{dt} = \underbrace{\overbrace{\sum_{j\neq i,\,p} TM_{j,i}^{p}}^{inputs} - \overbrace{\sum_{j\neq i,\,p} TM_{i,j}^{p}}^{outputs}}_{Transfer\ processes} \qquad (1)$$

where $[T]_i^{HTO}$ (x,t) is the HTO concentration of component i;

χ_i (x,t) is the density of component i (e.g. plant surface biomass);

$\chi_i [T]_i^{HTO}$ (t) is the HTO inventory of component i;

$TM_{i,j}^{p}$ (t) is the activity transfer process from component i to component j under process p.

A similar equation is used to estimate the temporal dynamics of non-exchangeable OBT concentration (i.e. $[T]_i^{OBT}$) of each component i considered.

In the case of chronic releases, the analytical solutions are calculated by solving the previous system of differential equations when the temporal derivatives are set to zero.

Input data

Type of Input Data

Type of Input	Input data
Radiological	Isotopic discrimination factors HTO dilution factors into plants through soil water HTO drinking water concentration Half-life related to plant growth dilution*
Ecological	Plant growth curves and associated parameters* Sowing, germination and harvest dates*
Physiological	Dry matter fractions Water equivalent factor Water fraction contributed to the diet by inhalation and skin absorption, and by food H metabolism
Trophic chain	Food and water ingestion rates

*Data required in the case of accidental releases only

Parameters

Symbol	Unit	Description
AIR		
H_a	kg m^{-3}	Absolute humidity
P	mm/month	Monthly precipitation
$[T]_{air}^{HTO}$	Bq m^{-3}	HTO concentration in air
$[T]_{precip}^{HTO}$	Bq L^{-1}	HTO concentration in precipitation
PLANT		
DI_{veg}	-	Isotopic discrimination factor
FD_f	-	HTO dilution factor in leaves by water coming from soil
FD_{veg}	-	HTO dilution factor in whole plant by water coming from soil
FE_{veg}	L kg^{-1} dw	Water equivalent factor
f_{veg}^{H2O}	L kg^{-1} fw	Average water fraction of plants

ANIMAL		
f_{ap}^{H20}	L kg^{-1} fw	Average water fraction of animal products
f_{ap}^{inhabs}	kg $kg^{-1}d^{-1}$	Water fraction contributed to the diet by inhalation and skin absorption
f_{ap}^{met}	kg kg^{-1}	Water fraction contributed to the diet by metabolism of hydrogen in food
FE_{ap}	L kg^{-1} dw	Water equivalent factor
$R_{water,ap}^{ing}$	L $animal^{-1}$ d^{-1}	Daily water ingestion rate
$R_{i,poa}^{ing}$	kg fw $animal^{-1}$ d^{-1}	Daily food ingestion rate
$[T]_{drink}^{HTO}$	Bq L^{-1}	HTO concentration in drinking water
SOIL		
D	m^{-2} s^{-1}	Diffusion coefficient of tritium into soil
F_{evap}^{HTO}	d^{-1}	Average evaporation rate
F_{transp}^{HTO}	kg m^{-2} d^{-1}	Average plant transpiration rate
f_{soil}^{H20}	L kg^{-1} fw	Water fraction of the sampled soils
H_{sol}	m	Soil layer depth
ρ_b	kg m^{-3}	Bulk density
ρ_v	kg m^{-3}	Saturation vapour mass at soil surface temperature
v_d	m s^{-1}	Dry deposition velocity

Parameter values and distributions

Parameter	Deterministic value	Distribution	Minimum	Maximum	Most likely value
AIR					
H_a , P , $[T]^{HTO}_{precip}$	cf. scenario description				
$[T]^{HTO}_{air}$	cf. scenario description	Uniform	-30%	+30%	
PLANT					
DI_{veg}	0.9	Triangular	0.7	1.1	0.9
FD_f	0.85	Uniform	0.8	0.9	
FD_{veg}	0.9	Uniform	0.85	0.95	
FE_{veg}	cf. scenario description	Uniform	-10%	+10%	
f^{H2O}_{veg}	cf. scenario description				
ANIMAL					
f^{H20}_{ap}	cf. scenario description				
f^{inhabs}_{ap} milk egg beef	0.021 0.036 0.028	Uniform	-10%	+10%	
f^{met}_{ap}	0.3	Uniform	0.2	0.4	
FE_{ap}	cf. scenario description	Uniform	-10%	+10%	
$R^{ing}_{i,poa}$	Total ingestion rates : Cows DF8 : 19 kg dw/d Cows DF11 : 10 kg dw/d Chicken F27 : 0.2 kg dw/d Ratios of feed components: cf. scenario	Uniform	-15%	+15%	
$R^{ing}_{water,ap}$	Cows DF8 : 75 L/d Cows DF11 : 75 L/d Chicken F27 : 0.3 L/d	Uniform	-15%	+15%	
$[T]^{HTO}_{drink}$	DF8 : 18.6 Bq/L DF11 : 21.1 Bq/L F27 : 24.3 Bq/L	— — Uniform	-10%	+10%	
SOIL					
D	$1x10^{-9}$ m^2/s				
F^{HTO}_{evap}	0.24 d^{-1}	Uniform	0.215	0.265	
F^{HTO}_{transp}	0.8 $L/m^2/d$	Uniform	0.7	0.9	
f^{H20}_{soil}	cf. scenario description				
H_{sol}	0.05 m				
ρ_b	1.08 g/cm^3				
ρ_v	0.015 kg/m^3				
v_d	0.001 m/s	Uniform	0.0005	0.0015	

Uncertainties

Uncertainties in the model predictions were determined using Latin Hypercube sampling of the distributions shown above in 1000 simulations.

TUM Model

Analytical procedures such as freeze drying and azeotropic distillation yield "buried" tritium as well as carbon bound tritium. The former amounts to the larger part according to the work of Baumgärtner (2005). Buried tritium is not relevant to long-term doses because it converts immediately by isotope exchange to HTO during digestion. Here, the Biochem Model calculates the amount of exchangeable tritium, including buried tritium. Carbon bound tritium, which is the tritium fraction that determines long-term dose, is obtained as the difference between OBT obtained experimentally and exchangeable tritium.

HTO Calculation

The model was driven by tritium concentrations in plants and animals rather than in air. Water inside plants or animals starts from the roots or the intestines and moves towards the leaves or the skin and kidneys. Therefore, it is assumed that the HTO concentration in plants and animals is equal to the mean of the HTO concentration in drinking water (Table 1) and in rainfall (Table 2) averaged over the 2-3 months prior to sampling; where the drinking water concentration was not available in July, the plant and animal concentrations were set equal to the average concentration in rain. Accordingly, the plant and animal HTO concentrations in July were determined to be 47, 30 and 446 Bq L^{-1} at the dairy farms, F27 and P2, respectively; for September, the corresponding values were 24.8, 22.7 and 447 Bq L^{-1}.

Table 1. Measured HTO concentrations drinking water in 2002 September

Compartment	DF8	DF11	F27	P2
Drinking water concentration (Bq L^{-1})	18.6	21.1	24.3	Not relevant

Table 2. Measured monthly HTO concentrations in precipitation.

Month	HTO Concentration in Precipitation (Bq L^{-1})		
	DF8	F27	P2
January	not available	not available	3670
February	not available	18	1350
March	not available	24	347
April	24	29	474
May	69	14	525
June	85	61	579
July	9	14	205
Mean April-July	**47**	**30**	**446**
August	49	19	442
September	13	22	452
Mean Aug-Sep	31	21	**447**
Mean of drinking water and precipitatation	**24.8**	**22.7**	

Buried Tritium

Buried tritium arises by triton-proton exchange during formation of biomolecules. We apply the fractionation factor $\alpha \approx 2$ as found in DNA. In the definition

$$\alpha = (Bq/ H_{ex})_{org}/(Bq/ H_{ex})_{aq} , \quad (1)$$

the tritium activity is denoted by Bq and the number of exchangeable hydrogen positions in the molecular unit by H_{ex}. Applying well-known definitions leads to

$$(Bq/ H_{ex})= (Bq/L)\ WE\ (M/ H_{ex}), \quad (2)$$

where WE is the water equivalent factor (L/kg) and M is the gram amount of the molecular unit. With $\alpha = 2$ we obtain

$$(Bq/L)_{org} = 2\cdot(Bq/L)_{aq}\cdot(18/2)\ (\ H_{ex}/M)_{\ org} / WE_{org}. \quad (3)$$

Assuming biomatter with buried hydrogen exists only in carbohydrates (CH_2O; cellulose, glycogen) and proteins, Eq. (3) becomes

$$(Bq/L)_{org} = 2\cdot(18/2)(Bq/L)_{aq}\cdot(18/2)\ [(\ H_{ex}/M)_{\ CH2O} + (H_{ex}/M)_{\ protein}]1000 / WE_{org}. \quad (4)$$

From the stoichiometry of carbohydrates $(C_6H_{10}O_5)_{\ n}$, M=162 and $(H_{ex}/M)_{\ CH2O} = 3/162 = 0.0185$. Taking account of the stoichiometric mean of 207 unrelated proteins (Klapper 1977), $(H_{ex}/M)_{protein} = 0.01676$. Plants are assumed to consist of carbohydrates only. The carbohydrate and protein contents of food are taken from the Nutrient Data Laboratory (www.nal.usda.gov-fnic-foodcomp-search).

References

Baumgärtner, F. 2005. Accumulative tritium transfer from water into biosystems. Proceedings of the 7th International Conference on Tritium Science and Technology. Baden-Baden, Germany, September 2004.

Klapper, M.H. 1977. The independent distribution of amino acids near neighbour pairs into polypeptides. Biochem. & Biophys. Research Comm. 78, 1018-1024.

FSA Model

The UK Food Standards Agency obtained predictions for the Pickering scenario using the Short-Term Atmospheric Release for H-3 (STAR H-3) model (Smith et al. 1995), which was developed by Intera Information Technologies (now part of Enviros Consulting). The model is implemented in the Amber software package (http://www.enviros.com/index.cfm?fuseaction=100&divisionId=6). STAR H-3 is a dynamic compartment model formulated in terms of a series of coupled first-order differential equations. Rate constants for the transfers between compartments were derived from consideration of the hydrogen inventories of the compartments and the hydrogen fluxes between them. Predictions for the Pickering scenario, which is an equilibrium situation, were obtained from the steady-state solution to the equations.

The model starts with a tritium concentration in air and consists of 6 compartments. These are:

Atmosphere: The air over an area of agricultural land in which the tritium concentration can be specified as a uniform or time varying concentration.

Soil in Root Zone: This contains hydrogen in soil water. All tritium in this zone is assumed to be in the form of tritiated water.

Plant Fast Turnover: This compartment represents the tritiated water and labile organically bound tritium in plant tissues

Plant Slow Turnover: This compartment represents the non-labile organically bound tritium within the plant tissues.

Animal Fast Turnover: The portion of an animal containing tritiated water and labile organically bound tritium

Animal Slow Turnover: The non-labile organically bound tritium within the animal.

The model can represent a range of crop and animal types.

The following transfers are represented within the model:

Transfer from Atmosphere to Soil in Root Zone: This transfer incorporates three components: HTO movement into the soil, water exchange between soil and atmosphere and wet deposition

Loss from Soil in Root Zone: Representing losses from the soil via evaporation and by transfer to deeper soil layers

Transfer from Soil in Root Zone to Plant Fast: Representing the uptake of water by plants. This process is driven by the net evapotranspiration of the plant.

Loss from Plant Fast: This process accounts for loss from the plant via evapotranspiration and exchange of tritiated water between plant and atmosphere.

Transfer from Atmosphere to Plant Fast: Representing the uptake of tritiated water by exchange with the atmosphere.

Transfer from Plant Fast to Plant Slow: Representing the incorporation of tritiated water and labile organically bound tritium into non-labile forms.

Transfer from Plant Slow to Plant Fast: The loss of non-labile tritium from plant tissues.

Transfer from Plant Fast and Slow to Animal Fast: Representing the consumption of crops by animals.

Transfer from Atmosphere to Animal Fast: This represents the intake by animals of tritium via inhalation.

Loss from Animal Fast: Representing the losses by excretion.

Transfer from Animal Fast to Animal Slow: The incorporation of tritiated water or labile organically bound tritium into animal tissues.

Transfer from Animal Slow to Animal Fast: Representing the loss of non-labile organically bound tritium from animal tissues.

The FSA calculations for the Pickering scenario were reviewed following the meeting of the EMRAS Tritium/C14 Working Group in Cardiff. This revealed that the default value for water content of air appropriate to UK conditions had been used instead of the value specified in the scenario. Use of the scenario specific value for this parameter would have decreased the FSA predicted concentrations by approximately 1/3.

Reference

Smith G.M., P.C. Robinson and M.J. Stenhouse. 1995. H-3 Foodchain Modelling Following Short Term Release to the Atmosphere. Intera Information Techologies.

SRA Model

The model calculations were based on the hypothesis that all the compartments are in equilibrium with each other. Soil tritium concentrations were estimated using the formula of Belot and others (BIOMASS 2003) that accounts for the contributions of both wet and dry deposition to the soil water concentration:

$$C_{sw} = \frac{v_d\ C_a\ + F_w}{v_e\ \rho_s\ + I_r},$$

where v_d is the transfer velocity of HTO from air to soil (m s^{-1}),
C_a is the tritium concentration in air (Bq m^{-3}),
F_w is the average flux density of tritium wet deposition (Bq m^{-2} s^{-1}),
v_e is the exchange velocity from soil to air (m s^{-1}),
ρ_s is the water concentration in air saturated at the soil surface temperature (L kg^{-1}), and
I_r is the infiltration rate of water through the root zone (m s^{-1}).

For the estimation of the free water tritium (FWT) concentration in plants, the following formula was introduced:

$$C_p = \alpha\left(\frac{C_a + I_W C_{sw} r}{\rho_s + \alpha I_W r}\right)$$

where C_p is the FWT concentration in the plant (Bq L^{-1}),
C_{sw} is the FWT concentration in soil water (Bq L^{-1}),
α is the isotope effect factor of HTO,
I_W is the average rainfall intensity (kg m^{-2} s^{-1}),
ρ_S is the saturated vapour density of the air in the atmosphere (kg m^{-3}), and
r is the exchange resistance for HTO and H_2O between the plant leaf and the atmosphere. Throughout the present calculations the value of r was assumed to be 67 s m^{-1}.

For all plant samples, including hay and haylage, rapid equilibration of plant FWT with atmospheric tritium vapour was assumed.

Since the water balance data for cows was not given, the daily free water intake was assumed to be 90 L d^{-1}. This value may be higher than the actual one. For tritium metabolism in lactating cows, the experimental results of Kirchmann et al. (1977, 1985) were referred to. By using a slightly modified version of their data, the ratio of the tritium specific activity in drinking water and the diet to that in milk was derived as follows:

Tritium source	Samples	Specific activity ratio
HTO	Milk dry matter	0.48
	Milk water	0.91
Tritiated feed	Milk dry matter	0.52
	Milk water	0.09

Determination of the confidence interval on the predictions was accomplished with some difficulty. The confidence range of the observed tritium concentrations in the field samples is unknown. The values of the various parameters used in the mathematical formulation depend on a number of other parameters and on the assumed environmental conditions under which the calculation was made. Under such conditions, it was necessary to make a basic assumption about the uncertainty of the driving parameters. Therefore, the standard deviations of the driving parameters were assumed to be 20% of the observed or assumed values.

References

BIOMASS. 2003. Modelling the environmental transport of tritium in the vicinity of long-term atmospheric and sub-surface sources. IAEA, Vienna.

Kirchmann, R., P. Charles, R. van Bruwaene and J. Remy. 1977. Distribution of tritium in the different organs of calves and pigs after ingestion of various tritiated feeds. Current Topics in Radiation Research, Q 12, 291.

Kirchmann, R., J. Remy, P. Charles and G. Koch. 1985. Distribution and incorporation of tritium in different organs of ruminants. IAEA-SM-172/81. International Atomic Energy Agency, Vienna.

GE Healthcare Model

The GE Healthcare model is a dynamic compartment model formulated in terms of a series of coupled first-order differential equations. Predictions for the Pickering scenario, which is an equilibrium situation, were obtained from the steady-state solution to the equations. The model is based on 1 kg of plant material, as this quantity can be easily amplified to the amount needed for consumption of crops within the critical group. The model starts with the tritium concentration in air and consists of four compartments representing the atmosphere, soil water, a plant fast compartment and a plant slow compartment. The plant fast compartment represents tissue free water inside the plant whilst the plant slow compartment represents the organic matter of the cells. It is assumed that these two compartments are in equilibrium within the plant. Transfer to animals was not modelled.

The following transfers are represented within the model:

- Transfer from the atmosphere to root zone soil water, including dry and wet deposition
- Loss from soil root zone by evaporation and transfer to deeper soil layers
- Transfer from root zone soil to the plant fast compartment, representing the uptake of water by plants.
- Transfer from the atmosphere to the plant fast compartment, representing the uptake of tritiated water by exchange with the atmosphere.
- Loss from the plant fast compartment, accounting for evapotranspiration and exchange of tritiated water between plant and atmosphere.
- Transfer from the plant fast compartment to the plant slow compartment, representing the incorporation of tritiated water and labile organically bound tritium into non-exchangeable forms.
- Transfer from the plant slow compartment to the plant fast compartment, accounting for the loss of non-exchangeable tritium from plant tissues.