EMRAS Tritium and C-14 Working Group

Soybean Scenario

Final Report

April 2008

1. SCENARIO DESCRIPTION

The soybean scenario addresses tritium absorption by soybean foliage and subsequent tritium behaviour in the plant. To provide data for model testing, soybean plants were exposed to elevated levels of airborne tritium in a glove box. The exposure was carried out acutely for one hour at various stages in the growth of the soybeans. The tritium behaviour in the plant body and pods was observed by sampling the various plant parts and determining the concentrations in them.

A total of six pots (SB1 to SB6) were tested, with the exposures occurring at different stages of growth. The sowing was made on May 22, flowering was observed on July 7, and harvest was done on October 5. The exposures were made on July 2, July 13, July 30, August 9, August 24 and September 17 for SB1 to SB6, respectively. SB1 and SB4 were sampled several times between exposure and harvest to measure the tritium concentrations of each plant part as a function of time. The other plants were sampled and analyzed twice, at the end of the exposure and at harvest. The surface soil of the pots was covered by vinyl paper during the exposure in order to prevent tritium from depositing to the soil. Following exposure, the plants were removed from the glove box and cultivated as usual outdoors.

Information on biomass growth rates, tritium concentrations in air in the glove box during the exposure, background tritium concentrations and meteorological conditions were given as part of the scenario. Modellers were asked to predict the following:

(i) HTO concentrations in the free water of the plant body and pods (tissue free water tritium – TFWT) in the SB1 and SB4 experiments at the times the plants were sampled;

(ii) the non-exchangeable OBT concentrations in the plant body and pods at harvest for each of the six experiments (SB1 to SB6); and,

(iii) the 95% confidence intervals on all predictions.

2. OBSERVATIONS

The observed concentrations corresponding to the requested predictions are shown in Tables 1, 2 and 3. The free water tritium and organically bound tritium concentrations were normalized by the mean activity of the air moisture in the glove box during the exposure. The normalized quantities make it easier to compare the trend of the calculations and observations across experiments, particularly for OBT, since the mean activities in air moisture in the glove box differed from experiment to experiment.

The observations have associated uncertainties that arose from sampling and counting; the supporting data (e.g. meteorological data and light intensity) also have related uncertainties. A Quantulus 1220 liquid scintillation counter (Wallac) was used to measure the tritium concentrations in the plant samples, with a counting error of about 10%. It is difficult to assign quantitative values to the other sources of uncertainty. The errors in the OBT measurements

would be higher than in the HTO concentrations because of difficulties in removing exchangeable OBT and combusting the dry matter. Some variability must be expected between plants, although this was kept to a minimum in the experiments by analyzing composite samples taken from a number of plants.

Table 1. Observed and normalized TFWT for SB1 experiment

Time and date	Time after exposure (hr)	TFWT (Bq/mL)	Normalized TFWT*
In the plant body (stem and leaves)			
10:40 July 2	0.2	9580	1.23E-01
11:30 July 2	1	1050	1.35E-02
July 3	24	3.92	5.05E-05
July 7	120	1.32	1.70E-05
July 16	336	0.33	4.25E-06
Aug. 10	936	0.11	1.42E-06
Sept. 7	1608	0.06	7.73E-07
Oct. 5	2280	0.06	7.73E-07
In the pods (shell and seeds)			
Aug. 10	936	0.21	2.70E-06
Sept. 7	1608	0.06	7.73E-07
Oct. 5	2280	0.06	7.73E-07

* Tissue free water tritium concentration in the plant divided by the average tritium concentration in air moisture during the exposure (7.77 E+04 Bq/mL for SB1)

Table 2. Observed and normalized TFWT concentrations for SB4 experiment

Time and date	Time after exposure (hr)	TFWT (Bq/mL)	Normalized TFWT*
In the plant body (stem and leaves)			
10:40 Aug. 9	0.2	7000	1.33E-01
11:30 Aug. 9	1	3200	6.08E-02
Aug. 10	24	25.9	4.92E-04
Aug. 14	120	2.1	3.99E-05
Aug. 23	336	0.8	1.52E-05
Sept. 10	768	0.27	5.13E-06
Oct. 5	1368	0.14	2.66E-06
In the pods (shell and seeds)			
10:40 Aug. 9	0.2	10500	1.99E-01
11:30 Aug. 9	1	8000	1.52E-01
Aug. 10	24	2700	5.13E-02
Aug. 14	120	63.5	1.21E-03
Aug. 23	336	1.49	2.83E-05
Sept. 10	768	0.84	1.59E-05
Oct. 5	1368	0.26	4.94E-06

*Tissue free water tritium concentration in the plant divided by the average tritium concentration in air moisture during the exposure (5.27 x 104 Bq/mL for SB4)

The TFWT concentrations in the plant body drop off much more quickly in experiment SB1 than in SB4, with values an order of magnitude or more lower between 24 and 120 hours post-exposure. This suggests that the tritium dynamics in the plants depend on the timing of the exposure relative to the growth stage of the plant. The difference in results may also be caused by differences in the growth rates of the plants and differences in the meteorological conditions that they experienced after the exposure.

The normalized TFWT concentration in the pods is higher than in the plant body for SB4, in particular from the time just after exposure to 120 hours elapsed. The TFWT in the plants decreased more rapidly than in the pods. This implies that the exchange rate of TFWT from the plant body to the air is higher than from the pods since most plant-to-air transfer occurs through the leaves. It also suggests that the transfer rate between the pods and the body is not high enough to preserve the equilibrium between the two parts of the plant. The TFWT concentration in the pods eventually dropped down to approximately the same level as that in the plant body, indicating that the TFWT comes into equilibrium throughout the plant after a sufficiently long time.

Table 3. Observed non-exchangeable organically bound tritium (OBT) concentration in plant parts at harvest for experiments SB1 to SB6

Case	Mean activity of air moisture during exposure (Bq/mL)	OBT concentration at harvest (Bq/mL)[1]							
		Body				Pods			
		Stem	Leaves	Avg.	Nor.avg.[2]	Shell	Seeds	Avg.	Nor.avg.[2]
SB1	7.77×10^4	18.0	14.0	16.0	2.06E-04	0.83	0.5	0.67	8.63E-06
SB2	1.47×10^5	59.8	50.8	55.3	3.75E-04	3.5	3.7	3.6	2.44E-05
SB3	1.14×10^5	37.8	17.7	27.8	2.44E-04	101.3	19.3	60.3	5.28E-04
SB4	5.27×10^4	19.8	8.8	14.3	2.71E-04	74.7	200.0	137.4	2.61E-03
SB5	9.19×10^4	44.3	13.5	28.9	3.14E-04	73.3	214.2	143.8	1.56E-03
SB6	1.37×10^5	180	19.5	99.8	7.28E-04	33.5	77.0	55.2	4.03E-04

[1] One gram of dry matter yields about 0.6 mL of combustion water
[2] Normalized OBT: average OBT concentration divided by the mean activity of air moisture

Table 3 shows the OBT concentrations at harvest for each experiment. For the plant body, separate concentrations are given for stems and leaves, as well as an average over the two compartments. For experiments SB3 through SB6, the concentrations in stems are quite different than in leaves, so the average must be treated with caution. A similar comment applies to shells and seeds and the average over the two compartments. However, the endpoint for the calculations was the plant body (stem + leaves) and the results submitted by most participants did not distinguish between shell and seeds. Thus, for simplicity, only two endpoints (plant body and pods) are considered here.

3. COMPARISON OF PREDICTIONS AND OBSERVATIONS

Twelve participants submitted predictions for the soybean scenario (Table 4), including KAERI. The scenario was not a blind test for KAERI, which provided the test data, but the KAERI model and predictions are included in the report since they provide insight into the results. Full descriptions of the models used to carry out the calculations are given in Appendix A.

Table 4. Participants in the soybean scenario

Participant	Affiliation	Designation used in the text
Phil Davis	AECL, Canada	AECL
Yves Belot	Consultant, France	Belot
Françoise Siclet	EDF, France	EDF
Wolfgang Raskob	FzK, Germany	FzK
Masahiro Saito	SRA, Japan	SRA
Kiriko Miyamoto	Japanet, Japan	Japanet
Hansoo Lee	KAERI, Korea	KAERI
Dan Galeriu	IFIN, Romania	IFIN
Alexei Golubev	VNIIEF, Russia	VNIIEF
Darren Cutts	FSA, United Kingdom	FSA
Paul Marks	GE HealthCare, United Kingdom	GE
Ring Peterson	LLNL, United States	LLNL

A generalized equation for the build-up of HTO concentration in the plant body during exposure can be expressed as an activity balance that includes tritium absorption for input and transpiration for output:

$$A\frac{dC_{pb}}{dt} = C_a - BC_{pb}, \qquad (1)$$

where C_{pb} and C_a are the HTO concentrations in the plant body and in air, respectively. A and B are the parameters required to sustain the proper activity balance between air and plant body. If the tritium concentration in air is constant, then the solution of Eq. (1) gives Belot's equation (Belot et al., 1979). Once the exposure is terminated and the chamber is opened, the air concentration to which the plants are exposed drops to natural background levels. Equation (1) is then employed again with the air concentration set to zero in order to predict the loss of tritium from the plant.

Most models take into account plant growth in calculating the HTO concentrations. Although plant growth rate data were given as part of the scenario, some modelers [AECL, FzK, IFIN] calculated it based on CO_2 assimilation or other approaches [VNIIEF] since there was considerable variability in the given data.

The transfer of HTO from the plant body to other organs was modeled as an instantaneous equilibrium with different partitioning factors for shells and seeds [AECL, SRA, FSA, GE, KAERI, LLNL] or with a single factor for the pods as a whole [Belot, FzK].

In some models, OBT formation was treated as an equilibrium, with appropriate parameters relating the OBT and HTO concentrations in each organ [Belot, Japanet]. Some codes allowed the OBT concentration to be diluted by new, uncontaminated growth [AECL, IFIN, LLNL, VNIIEF]. Other models related OBT formation to HTO concentrations using forward and backward transfer rates for the plant body and forward transfer rates only for the pods [FSA, FzK, KAERI, SRA]. In these cases, plant growth was incorporated into the calculations by adding the plant growth balance equation. Most models also account for the reverse transfer from OBT to HTO by introducing rate constants for the plant body and the pods [SRA, FSA, GE, KAERI, LLNL]. In this way the models describe the different rates of decrease of tritium in the plant body and the pods.

One of the participants in the Working Group, Franz Baumgartner of the Technical University of Munich, contributed a paper discussing a conceptual model that describes tritium transfer from water to biomolecules by energy balance between hydrogen isotopes, i.e. by minimizing the free energy of the isotopes. Although no results were submitted for this model, it is described in Appendix A as well.

Four participants [LLNL, Japanet, AECL and IFIN] submitted estimates of the 95% confidence intervals on the predicted concentrations. LLNL carried out a numerical Monte Carlo uncertainty analysis using input parameters with normal, triangular or uniform distributions. The uncertainties estimated by Japanet for the TFWT concentrations were based on a 10% variation in the rate constant of HTO loss from the plant; for OBT, the uncertainties were based on the standard deviations of the mean HTO concentration in air moisture during the exposure. The AECL estimates were based on an uncertainty analysis of UFOTRI, a code similar to ETMOD, for a scenario from BIOMOVS II that was similar to the soybean scenario (BIOMOVS II, 1996). The uncertainties are not shown in the figures to prevent them from becoming too busy, but are discussed in Section 3.4.

3.1 HTO Concentrations for SB1

Normalized HTO concentrations in the plant body and in the pods for experiment SB1 are shown as a function of time in Figures 1 and 2, respectively. The predictions of most participants for the plant body lay close together in the early part of the experiment (up to 1 hr after the exposure). All predictions lay above the observed data but by less than an order of magnitude in most cases. The reason for this is not clear. When the exposure was finished, a fan was used to remove the tritium from the glove box. This may have caused extreme mixing that facilitated the removal of tritium from the plant body. Another reason for low uptake by the plants may have been the high temperatures in the chamber, which may have affected the behavior of the stomata. Alternatively, the models themselves may have been in error, with uptake rates that were too high or initial loss rates that were too low.

The predictions of the various models diverged significantly after 1 hour. EDF, FzK and IFIN predicted the observations closely in the beginning, but the calculations showed a sudden change after 1 hr, resulting in predictions that were lower than the observations at later times. GE, KAERI and SRA predictions were higher than the measurements through the entire observation time. The predictions as a whole show no obvious bias but the results of individual models are more than three orders of magnitude different from the observations in some cases.

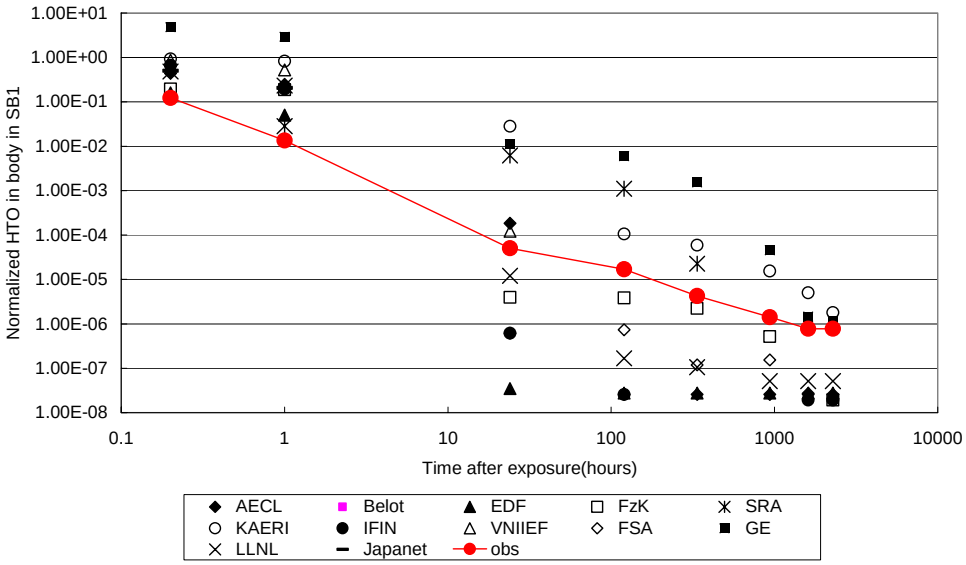

Fig. 1. Predicted and observed normalized HTO concentration in plant body in SB1

The predictions of normalized HTO concentration in the pods in SB1 (Figure 2) varied from 10^{-4} to 10^{-9}, whereas the observations lay in the range of 10^{-6}. According to the observations, the HTO concentrations were roughly the same in all parts of the plant at these times. Most of the models reproduced this observation and thus over- or underestimated the concentration in the pods to the same extent that they over- or underestimated the concentrations in the plant body. LLNL assumed that the pods were not growing at the time of exposure for SB1 and that HTO in the pods when they started to grow equaled the HTO in the leaves. This assumption accounted for the low predictions of LLNL in Fig. 2.

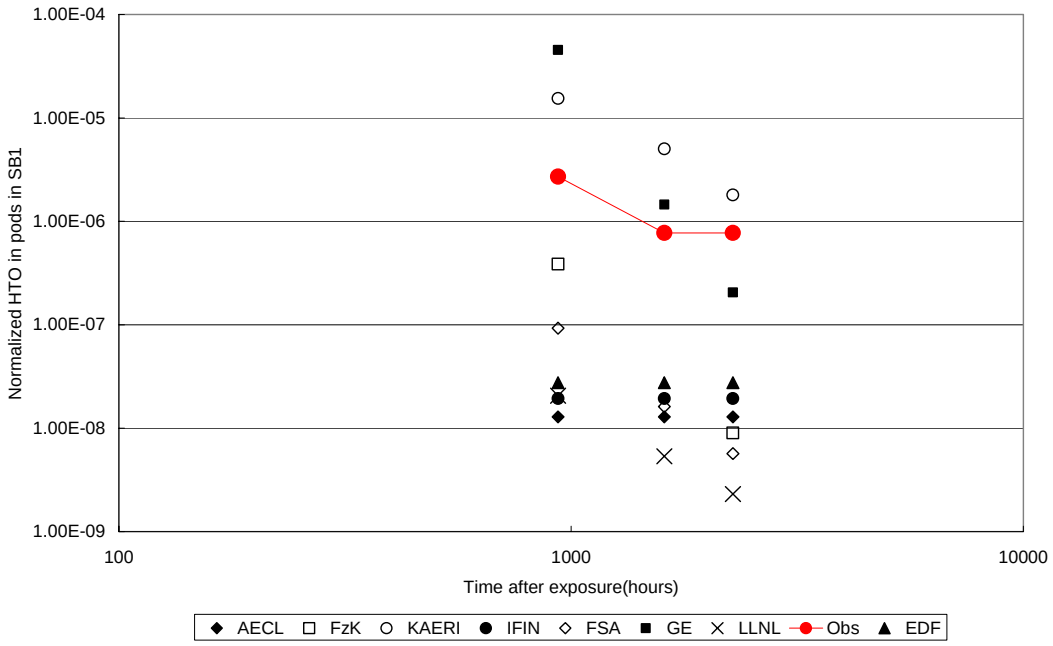

Fig. 2. Predicted and observed normalized HTO concentration in pods in SB1

3.2 HTO Concentrations for SB4

The predicted HTO concentrations in the plant body for experiment SB4 (Figure 3) show the same patterns as the predictions for SB1. The results of all participants are fairly closely grouped and higher than the observations at the beginning of the experiment, but after 24 hours they become distributed over a range of 10^5, with some overestimates and some underestimates. As was the case for SB1, the discrepancy could be explained by the use of uptake rates that were too high or initial loss rates that were too low.

The predictions and observations of normalized HTO concentration in the pods for SB4 are shown in Fig. 4. At the beginning of the experiment, the predictions range over a factor of 100 and bracket the observations. At later times, the predictions range over five orders of magnitude and tend to underestimate the observations. Most of the models are unable to account for the relatively long residence time of HTO in the pods.

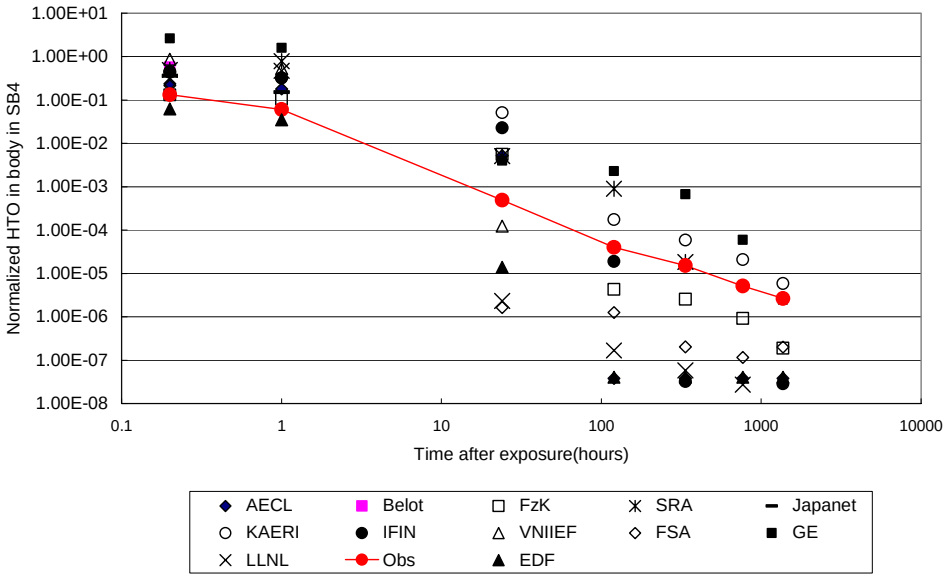

Fig. 3. Predicted and observed normalized HTO concentration in plant body in SB4

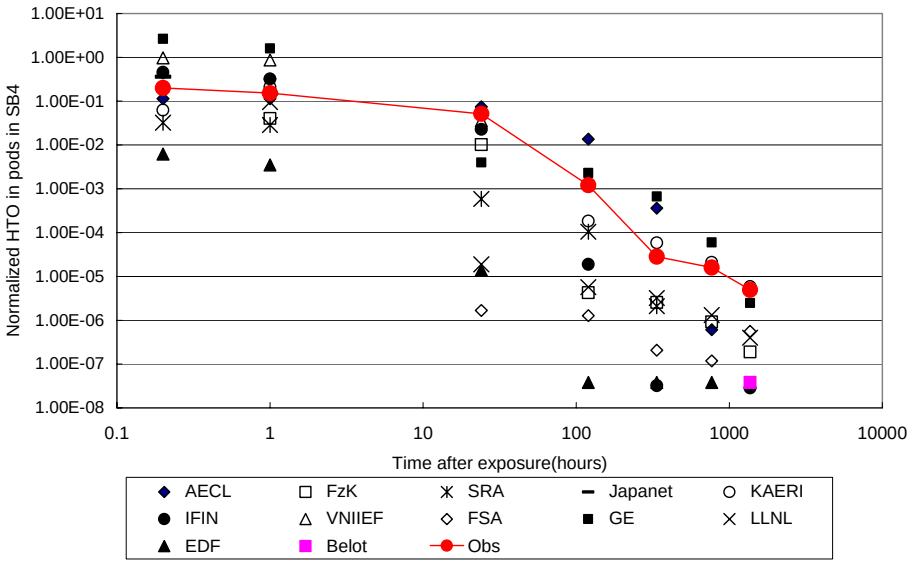

Fig. 4 Predicted and observed normalized HTO concentration in pods in SB4

The HTO concentrations in all parts of the plants at harvest were about 2 orders of magnitude higher than the levels expected of plants growing in an environment with an average air concentration of 0.04 Bq m^{-3}. There are two possible explanations for this:

(i) The HTO concentrations are maintained at a relatively high level by the slow breakdown of OBT in the plant.

(ii) The high concentrations may occur as the result of a reverse transfer of HTO from stem or roots to leaves and pods. In the pre-fruiting period, part of the HTO is transferred to stem and roots. At the fruiting period, when the HTO in the leaves and pods is almost exhausted by translocation or transpiration, the HTO in the stem or roots is recycled to the leaves and pods, maintaining the concentration at the residual level.

In theory, elevated concentrations could also be maintained by root uptake if the soil was contaminated during exposure. However, this is believed to be unlikely because of the care taken in applying the vinyl covering to the soil and the post-exposure dilution of soil water concentrations with clean irrigation water.

3.3 OBT Concentrations for SB1 to SB6

The predictions and observations for OBT concentrations for experiments SB1 to SB6 are plotted in Figs. 5 and 6 for the plant body and the pods, respectively. The observed concentrations in the plant body increased slightly as the time between exposure and harvest decreased. This likely reflects the fact that the OBT in plants exposed at later times had less time to breakdown and was less subject to dilution with new, uncontaminated dry matter production. The predictions of most models were fairly constant with time and followed the observed tendency well, even though the absolute values were dispersed over two orders of magnitude. The predictions tended to underestimate the observations, more so for the first three experiments than for the last three. The AECL model under-predicted severely for SB3-SB6. This model assumed that all OBT formed in the leaves after flowering was translocated to the pods and set the leaf concentration to background levels. This assumption is not supported by the observations.

The OBT concentration in the pods is crucial for estimating the effects of contaminated foodstuffs in the diet. This concentration initially increased as the time between exposure and harvest decreased, reaching a maximum for SB4 when the plants were growing very actively (Fig. 6). Concentrations dropped off as the exposure took place closer to harvest, perhaps because the translocation rate to the grain decreased as the grain became riper. Moreover, the leaves started to fall in the later experiments, reducing the amount of tritium absorption through the leaves and making less tritium available for OBT formation. The predictions of some models captured this variation well while others remained almost constant or increased only slightly with exposure time. The very low predictions by the AECL, LLNL, EDF and IFIN models for SB1, in which the pods had not yet started to form at the time of exposure, occurred because the HTO concentrations in these models drop off very quickly with time and were essentially negligible when the pods began to form. The low concentration predicted by AECL for SB2 has a similar explanation.

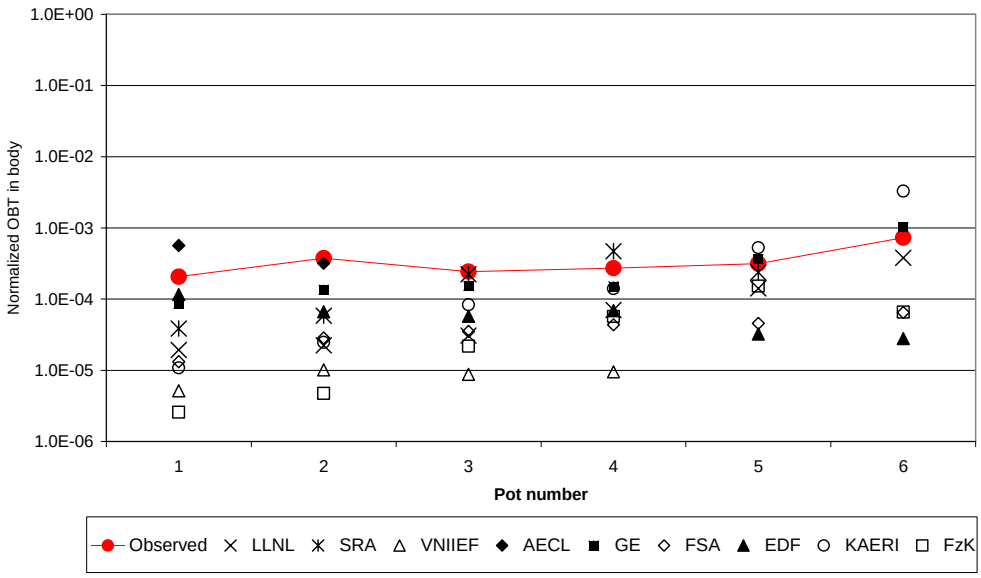

Fig. 5. Predicted and observed OBT concentrations in the plant body at harvest (average of stem and leaves). AECL predictions for SB3 to SB6 were ~ 10^{-8} and are not plotted.

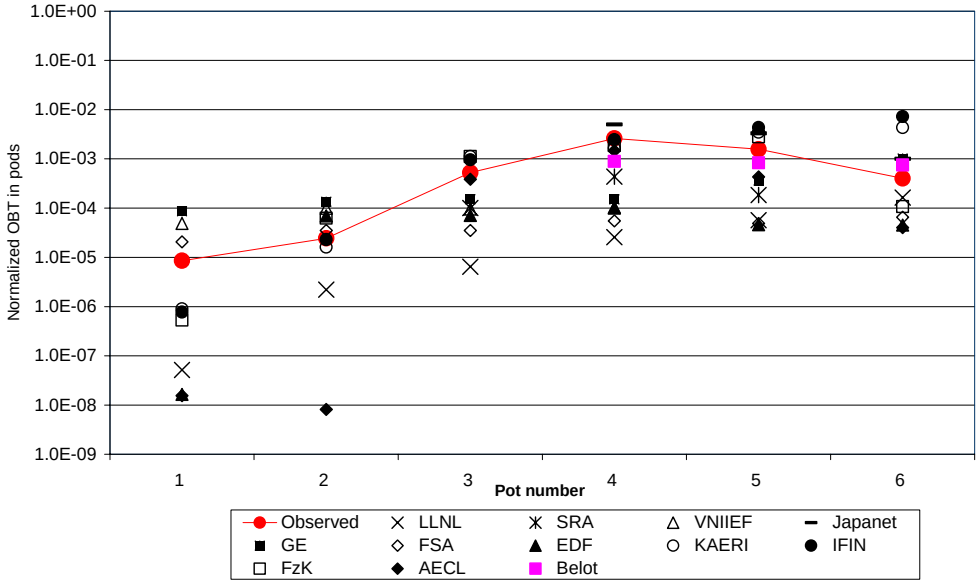

Fig. 6. Predicted and observed OBT concentrations in the pods at harvest (average of shell and seeds)

The amount of OBT formed in a plant depends on the time-integrated HTO concentration in its leaves rather than on the instantaneous HTO concentration at any given time. Accordingly, ratios of OBT at harvest to the HTO concentration in the plant body integrated over the first 24 hours following exposure were calculated for each model and compared with the observations in Fig. 7 to provide insight into the behaviour of the models. The calculated ratios were lower than the observed ratios for both SB1 and SB4, whether the ratio was based on the OBT in the plant body or in the pods. The models generate a significantly smaller amount of OBT per unit time-integrated HTO concentration than the plants produce in reality. The models that overpredict the OBT concentration in the plants do so only because they overpredict the initial time-integrated HTO concentration. This is of concern since it is the OBT concentration that is important in determining dose.

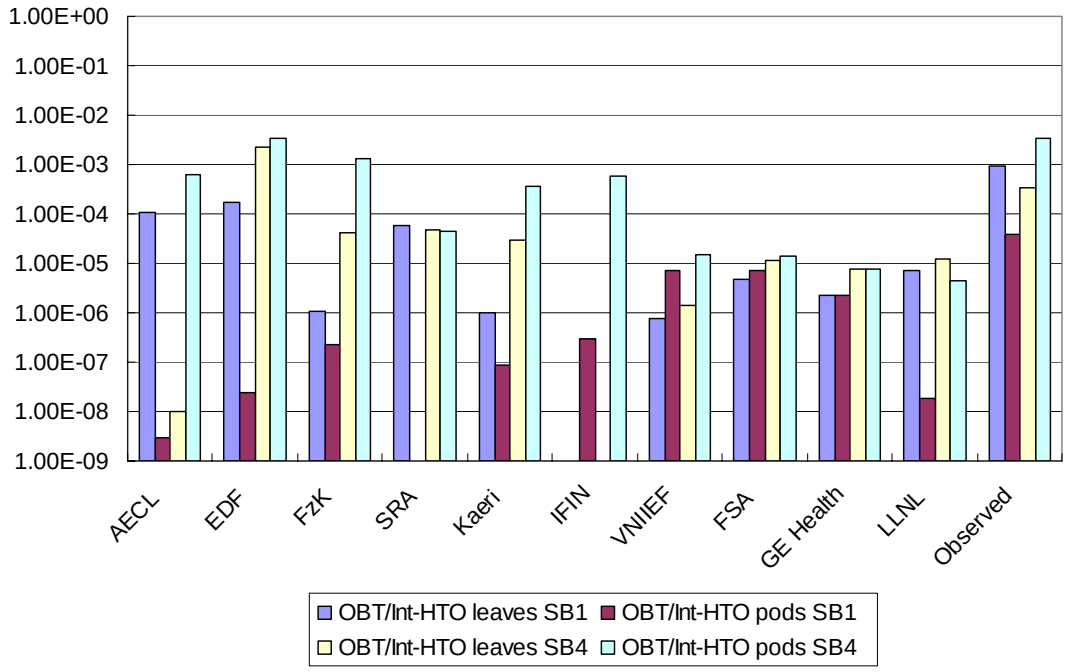

Fig. 7. The ratio of OBT in plant parts to the HTO concentration integrated for 24 hrs

In the SB1 experiment, the observed OBT/time-integrated HTO ratio was higher in the leaves than in the pods. Most of the OBT remained in the plant body since the exposure was carried out before flowering. All models except VNIIEF and FSA reproduced this behaviour. In contrast, for SB4, the ratio was higher in the pods than in the leaves since most of the OBT was translocated to the pods as this exposure was conducted during the active fruiting period. Again, all of the models with the exception of SRA and LLNL were able to simulate this result.

3.4 Uncertainty Analysis

Four participants (LLNL, Japanet, AECL and IFIN) calculated the uncertainties on their predictions and arrived at very different conclusions. The Japanet estimates were very low,

with the upper and lower confidence limits lying just 5% from the best estimate. In the case of AECL, the 95% confidence interval (the 97.5th percentile divided by the 2.5th percentile) was estimated to cover a factor of 10 for TFWT concentrations in the plant body and a factor of 4 for OBT concentrations in pods at harvest. The uncertainties were largest for concentrations predicted immediately after exposure and decreased slightly thereafter. The confidence intervals on TFWT estimated by both LLNL and IFIN depended strongly on the time after exposure. The intervals were relatively low (~ a factor of 2 - 5) immediately after exposure and again at very long times. At intermediate times, the confidence intervals were larger, with values as high as a factor of 100 at t = 24 hours for SB1. For OBT concentrations in the pods, the LLNL and IFIN uncertainties depended on the growth stage of the plant at the time of exposure but the confidence intervals generally lay between factors of 10 and 30.

Given the large variation in the confidence intervals estimated by the various participants, no definitive conclusions can be drawn regarding the uncertainties in the model predictions. Ideally, the confidence intervals would take into account the uncertainties in the HTO concentrations in air moisture used to drive the models; in the various transfer parameters required by the models; in specifying the growth rates of the plants; and in the structure of the models themselves. The best estimate of uncertainty can perhaps be obtained from an overall assessment of the scatter in the predictions submitted by the participants. These suggest that the confidence intervals on TFWT concentrations are about a factor of 10 shortly after exposure, increasing to a factor of 1000 or more at later times. The greater uncertainty at longer times is probably due to differences in how the models treat residual HTO due to sequestration or the breakdown of OBT. The confidence intervals on the OBT concentrations in the pods at harvest are about a factor of 100 or less, with the experiments at later stages of plant growth having the lower uncertainties. The confidence intervals are generally smaller for OBT than for TFWT, reflecting the fact that HTO varies rapidly over time whereas OBT integrates.

4. SUMMARY AND CONCLUSIONS

The soybean scenario tested the rate of tritium transfer between air and plant and the rate of OBT formation. Six experiments were carried out using a glove box to expose plants at different stages of growth. Two of the experiments were designed to study the time-dependent HTO concentration in various parts of the soybeans. After the exposure, the soybean parts were sampled with time to quantify the rate of transfer from leaves to other plant parts or to the air as time elapsed. In all experiments, the OBT concentrations were measured in the plants parts at harvest.

The observations were normalized to the air moisture concentrations in the exposure chamber so that a more meaningful comparison of the results could be made across experiments. The HTO concentrations in the pods were higher than in the plant body in the early period after the exposure, since the transpiration to air from the pods was lower than from the plant body. The OBT concentration in the pods at harvest initially increased as the time between exposure and harvest decreased, with the highest concentration occurring for the exposure time closest to the active fruiting period. For exposure at later growth stages, the OBT concentration decreased

slightly due to low translocation rates or lesser numbers of leaves supplying tritium from the air.

Twelve participants submitted predictions and one submitted a theoretical paper concerning tritium transfer from water to biota. The models for tritium transfer from air to leaves were generally based on an activity balance, yielding the HTO concentrations in the plant body or pods. Some participants used equilibrium assumptions to calculate OBT concentrations based on the HTO levels whereas others used compartment models to simulate HTO and OBT concentrations simultaneously and time-dependently.

The predictions of HTO concentrations in the plant body for SB1 were higher than the observations in the first hour after exposure. This might be due to the vigorous mixing used to remove the tritium from the glove box after the experiment, which may have accelerated the transfer of tritium from the leaves to the air. Alternatively, the discrepancy may be explained by the use of incorrect values for the transfer parameters in the models. After an hour, the predictions dispersed widely but followed the observed trend to decreasing HTO concentrations with time. Predictions for the normalized HTO concentration in the pods in SB1 ranged widely again but scattered around the observations.

The results for the HTO concentration in the plant body for SB4 were similar to those for SB1. For both experiments, the concentrations at harvest were well above background levels. There are a number of possible explanations for this observation, but a definitive conclusion must await additional experimental evidence. The generally poor predictions at longer times are of no practical significance because the concentrations are extremely low at this point. The models are all conservative for the first few hours, the period that is important for the production of OBT.

OBT concentration is the essential information necessary for assessing ingestion doses. Most participants were able to reproduce the slight increase in the normalized OBT concentrations that was observed in the plant body at harvest as the time between exposure and harvest decreased. Similarly, most participants were able to simulate the variation in the normalized OBT concentration in the pods with exposure time by considering the plant growth rate in their models. However, most of the models underestimated the observed OBT concentrations, and the scatter in the predictions, while less than that for HTO, remained substantial. The models do not produce as much OBT per unit time-integrated HTO concentration as the plants were observed to produce. The models would have underestimated the OBT concentration by more than they did if they had not overpredicted the initial time-integrated HTO concentration.

Four participants carried out uncertainty analyses and reported the 95% confidence interval on their predictions. These differed substantially and no definitive conclusions can be drawn regarding the uncertainties of the models. An overall assessment of the scatter in the predictions suggests that the confidence intervals on TFWT concentrations are about a factor of 10 shortly after exposure, increasing to a factor of 1000 or more at later times. These large uncertainties are of little significance because the concentrations themselves are very low at these times. The confidence intervals on the OBT concentrations in the pods at harvest are

about a factor of 100 or less, with the experiments at later stages of plant growth having the lower uncertainties.

5. REFERENCES

Belot , Y., Gauthier, D., Camus, H., Caput, C., “Prediction of the flux of tritiated water from air to plant leaves”, Health Physics 37, 575 (1979)

BIOMOVS II, Tritium in the food chain: intercomparison of model predictions of contamination in soil, crops, milk and beef after a short exposure to tritiated water vapor in air, Technical Report No.8, Swedish Radiation Protection Institute, Stockholm, Sweden (1996)

Appendix A

Model Descriptions

AECL Model Description and Discussion of Results

1. INTRODUCTION

Model Name: ETMOD (Environmental Tritium MODel)

Model Purpose: ETMOD was developed as a research code but has been used as an assessment tool to predict the consequences of accidental tritium releases to the atmosphere from tritium-handling facilities. It is intended to be realistic.

Type of Model: ETMOD is a dynamic, process-oriented model.

Compartments Considered: Air, soil, plants and animals.

Transport Processes Considered: ETMOD covers many transport and exposure pathways including atmospheric dispersion, dry and wet deposition to soil, migration in soil, re-emission from soil, and transfer to vegetation, animals and animal products. It can handle releases of either tritium gas (HT) or tritiated water vapour (HTO) and addresses organically bound tritium (OBT) formation in plants.

Endpoints: Final endpoints are ingestion and inhalation (including skin absorption) doses to humans. Intermediate endpoints include tritium concentrations in the various environmental compartments.

References: Russell and Ogram 1992, Thompson et al. 1992.

2. KEY ASSUMPTIONS AND MODELLING APPROACHES

5.1 Tritium transfer between air and plants

The exchange of tritium between air and plants is modeled as a diffusion process with the transfer driven by the concentration gradient between air and leaf:

$$\frac{dC_{pw}}{dt} = \frac{V_{ex}}{M_w}(C_a - \gamma\ h\ C_{pw}), \qquad (1)$$

where C_{pw} is the HTO concentration per unit mass of plant water (Bq kg^{-1}),
V_{ex} is an exchange velocity (m s^{-1}),
M_w is the mass of plant water per unit ground surface area (kg m^{-2}),
C_a is the HTO concentration in air (Bq m^{-3}),
γ is the ratio of the vapour pressure of HTO to H_2O (0.91), and
h is the saturation humidity at leaf temperature (assumed equal to air temperature) (kg m^{-3}).

Eq. (1) describes both deposition to the plant and emission from the plant, depending on the sign of the term in brackets on the right side of the equation. The exchange velocity is calculated using the multiple resistance approach, taking into account the aerodynamic resistance to transfer through the air, the boundary-layer resistance through the laminar sublayer very close to the plant surface, and the stomatal or canopy resistance through the surface of the plant itself. The aerodynamic and boundary-layer resistances are calculated using meteorological data for the current time step. The stomatal resistance is taken from Wesley (1989), who provides values as a function of season and land use. These numbers were modified to account for the values of solar radiation and surface temperature observed at the time of the calculation. The HTO concentrations predicted by Eq. (1) are assumed to apply to all aqueous compartments of the plant.

5.2 Dry Matter Production

Gross photosynthesis rates are calculated using the CO_2 consumption model (Weir et al. 1984, Sellers 1985, Mitchell et al. 1991, Pinder et al. 1988) and depend on air temperature, the resistance to CO_2 uptake by the plant and the photosynthetically active radiation reaching the plant, which in turn depends on leaf area index. The production rate of dry matter is based on net photosynthesis (the difference between gross photosynthesis and respiration), taking into account both growth and maintenance respiration. Plant dry mass is updated using the dry matter produced in the time step. The wet vegetation mass is then calculated from the dry mass and the fractional water content, which is assumed to remain constant as the plant grows. The calculation stops when a pre-specified plant mass or harvest time is reached.

5.3 OBT Formation

The dry matter produced at a given time is assumed to have a T/H ratio equal to 0.6 times the T/H ratio in the plant water that takes part in the photosynthesis at that time. OBT concentrations following exposure decrease due to dilution with new uncontaminated dry matter. ETMOD does not account for the slow conversion of OBT to HTO in plants due to metabolic processes. OBT concentrations calculated in this way are assumed to apply to all dry matter in the plant.

5.4 Translocation

ETMOD can handle four types of crops (pasture, leafy vegetables, root vegetables and grain). In each case, the plant is treated as a single compartment with uniform concentrations throughout. This means that translocation between different parts of the plant must be addressed outside ETMOD. For this scenario, the soybeans were treated as leafy vegetables, and simple conceptual and mathematical models were used to simulate the transfer of tritium between the soybean leaves and the pods. The following assumptions were made:

- The HTO concentration in the pods at the end of the exposure is half the concentration in the leaves. This concentration is reduced through dilution as the plant grows and through losses to the air, with a half time of 2 days.
- Once the leaves and stems are fully grown, all new OBT produced is translocated to the pods. This OBT is distributed in proportion to the stage of development of the plant, with more OBT going to the faster growing of the shells and seeds.

- OBT concentrations in the shells and seeds were calculated by mixing the amount of OBT translocated into the observed dry weight of these compartments at harvest.

3. PARAMETER VALUES

ETMOD contains a large number of parameters for which values must be specified. These include fixed values for parameters relating to site characteristics, soil properties, plant properties, weather data, dosimetry and the scenario in question. In addition, hourly values of such meteorological parameters as wind speed, air temperature, humidity, cloud cover and precipitation must be entered, together with time-dependent release rates.

4. MODEL UNCERTAINTIES

A rigorous uncertainty analysis of ETMOD has not been undertaken. However, the 95% confidence interval is estimated to cover a factor of 10 for HTO concentrations in the plant body and a factor of 4 for OBT concentrations in the pods at harvest, based on results of an uncertainty analysis for UFOTRI, a code similar to ETMOD, for a scenario from BIOMOVS II that was similar to the soybean scenario (Galeriu et al. 1995). These estimates reflect the uncertainty due to parameter values only and do not include uncertainties due to model structure. The results for UFOTRI suggest that the uncertainties are largest for concentrations predicted immediately after exposure and decrease slightly thereafter.

5. APPLICATION OF ETMOD TO THE SOYBEAN SCENARIO

Each simulation began by setting the fresh weights of the plants and their water contents equal to the values observed at the beginning of the exposure (Table 1). The water contents were assumed constant with time as the plant grew. The air concentration in the model was set equal to the average concentration observed in the chamber (Table 1) for one hour and then decreased to zero. It was found that no plant dry matter was produced when the model was run with the air temperature observed in the chamber. Photosynthesis is strongly temperature-dependent and the model in ETMOD assumes that little dry matter is formed for temperatures above 40^{o}C. Accordingly, the temperatures during exposure were arbitrarily set equal to the mean of the temperatures inside and outside the chamber. In all hours after the exposure, the observed temperatures were reduced by 4.2^{o}C, the difference between temperatures in Korea and Canada, to better reflect the Canadian conditions for which ETMOD was developed. The meteorological data supplied with the scenario were filled in so that values for all parameters were available every hour. It was assumed that photosynthetically active radiation equals one-half incoming solar radiation and that the water equivalent factor for soybeans is 0.57. Time steps varied from 0.01 hours for the first 48 hours of each simulation to 0.1 hours for the remainder of the runs.

The leaf area index (LAI) was not calculated in the model but rather was pre-defined at the outset of the run based on information provided in the scenario description.

Predicted plant concentrations were not allowed to drop below the background values that would be expected for a plant growing in an environment with an average air concentration of 0.04 Bq/m^3.

Table 1. Air concentrations in the chamber and plant water contents and fresh weights.

Experiment	Air concentration (Bq/L)	Plant water content (%)	Initial plant fresh weight (kg/m^2)
SB1	8.42×10^7	82.0	0.96
SB2	1.59×10^8	78.7	1.22
SB3	1.24×10^8	73.3	2.73
SB4	5.71×10^7	68.7	2.02
SB5	9.96×10^7	68.3	4.17
SB6	1.49×10^8	67.5	3.59

6. DISCUSSION OF AECL RESULTS

6.1 Exchange Velocities, Fluxes, Dry Matter Production Rates and Plant Masses

Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for runs SB1 through SB6 are shown in Tables 2-7. The exchange velocities fluctuate according to the current meteorological conditions, with most values lying between 2×10^{-3} and 8×10^{-3} m/s for these daytime conditions. The values decrease by about an order of magnitude toward the end of each run because fall values for the stomatal resistance were used rather than summer values. The HTO flux is directed into the plant at the start of each exposure but reverses as soon as the exposure ends and quickly goes to zero or very small values as the HTO diffuses out of the leaves. The predicted plant mass increases throughout the simulation period for SB1 and SB2, goes through a maximum for SB3 and SB4, and decreases uniformly for SB5 and SB6. In each case, the predicted mass at harvest is substantially smaller than the observed mass, by more than a factor of 3 in the case of SB2. Clearly ETMOD is underestimating the dry matter production rate. The leaf area index does not always increase and decrease in phase with the plant mass since the former is an imposed quantity and the latter is calculated. The assumption that the water content of the plants stays constant with time appears to be good for SB4 and SB5, but in all other cases the observed water contents decreased significantly over the study period.

Table 2. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB1. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	4.93×10^{-3}	-1.22×10^{4}	0.961	2.93	2.35×10^{-7}
0.2	11	7.61×10^{-3}	5.24×10^{3}	0.967	2.93	2.66×10^{-7}
1	12	7.61×10^{-3}	3.03×10^{3}	0.971	2.93	2.66×10^{-7}
24	11	6.92×10^{-3}	2.12	1.01	3.02	1.67×10^{-7}
120	11	6.95×10^{-3}	0	1.18	3.39	1.91×10^{-7}
336	11	5.97×10^{-3}	0	1.41	4.21	8.99×10^{-8}
936	11	6.21×10^{-3}	0	1.94	6.50	7.60×10^{-8}
1608	11	7.44×10^{-4}	0	2.31	5.70	6.48×10^{-8}
2280	11	6.44×10^{-4}	0	2.37	3.00	5.48×10^{-8}

* from plant to air

Table 3. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB2. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	5.79×10^{-3}	-2.16×10^{4}	1.22	3.94	1.47×10^{-7}
0.2	11	2.06×10^{-3}	2.05×10^{3}	1.22	3.94	1.84×10^{-8}
1	12	2.06×10^{-3}	1.72×10^{3}	1.22	3.94	1.84×10^{-8}
24	11	2.06×10^{-3}	1.16×10^{2}	1.23	4.03	1.78×10^{-8}
120	11	6.90×10^{-3}	6.92×10^{-6}	1.29	4.39	1.60×10^{-7}
336	11	6.35×10^{-3}	0	1.46	5.22	1.13×10^{-7}
768	11	5.42×10^{-3}	0	1.72	6.50	4.83×10^{-8}
936	11	6.68×10^{-3}	0	1.82	6.50	1.04×10^{-7}
1368	11	6.81×10^{-4}	0	2.01	5.60	5.44×10^{-8}
1608	11	6.30×10^{-4}	0	2.03	4.60	5.13×10^{-8}
2016	11	6.94×10^{-4}	0	2.05	3.00	5.45×10^{-8}

* from plant to air

Table 4. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB3. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	4.21×10^{-3}	-1.72×10^{4}	2.73	5.49	7.46×10^{-8}
0.2	11	6.78×10^{-3}	7.32×10^{3}	2.73	5.49	8.69×10^{-8}
1	12	6.78×10^{-3}	4.36×10^{3}	2.73	5.49	8.69×10^{-8}
24	11	6.27×10^{-3}	3.20×10^{1}	2.72	5.58	3.85×10^{-8}
120	11	5.68×10^{-3}	4.02×10^{-1}	2.73	5.95	3.75×10^{-8}
336	11	6.13×10^{-3}	1.95×10^{-1}	2.73	6.50	1.86×10^{-8}
768	11	6.66×10^{-3}	2.43×10^{-1}	2.76	6.40	6.31×10^{-8}
936	11	7.44×10^{-4}	3.52×10^{-2}	2.74	5.70	3.94×10^{-8}
1368	11	6.89×10^{-4}	4.02×10^{-2}	2.63	3.90	3.76×10^{-8}
1608	11	6.94×10^{-4}	1.99×10^{-2}	2.57	3.00	3.40×10^{-8}

* from plant to air

Table 5. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB4. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	3.64×10^{-3}	-5.99×10^{3}	2.02	6.41	1.47×10^{-7}
0.2	11	7.39×10^{-3}	2.47×10^{3}	2.02	6.41	1.51×10^{-7}
1	12	7.39×10^{-3}	1.53×10^{3}	2.02	6.41	1.51×10^{-7}
24	11	7.09×10^{-3}	4.26	2.02	6.50	1.21×10^{-7}
120	11	5.42×10^{-3}	0	2.02	6.50	2.58×10^{-8}
336	11	6.62×10^{-3}	0	2.05	6.50	7.11×10^{-8}
768	11	5.22×10^{-4}	0	2.06	5.40	1.47×10^{-8}
936	11	5.36×10^{-4}	0	2.03	4.70	2.19×10^{-8}
1368	11	6.44×10^{-4}	0	1.97	3.00	3.89×10^{-8}

* from plant to air

Table 6. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB5. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	5.48×10^{-3}	-1.39×10^{4}	4.17	6.50	5.51×10^{-8}
0.2	11	7.39×10^{-3}	5.63×10^{4}	4.17	6.50	9.43×10^{-8}
1	12	7.39×10^{-3}	3.40×10^{3}	4.17	6.50	9.43×10^{-8}
24	11	5.61×10^{-3}	9.16	4.16	6.50	2.14×10^{-8}
120	11	6.69×10^{-3}	0	4.11	6.50	1.36×10^{-8}
336	11	7.44×10^{-4}	0	3.97	5.70	-8.50×10^{-8}
768	11	6.89×10^{-4}	0	3.66	3.90	-4.30×10^{-8}
1008	11	6.94×10^{-4}	0	3.52	3.00	-5.18×10^{-8}

* from plant to air

Table 7. Time-dependent ETMOD predictions of exchange velocity, HTO flux from leaf to air, plant mass and dry matter production rate for run SB6. The imposed values of leaf area index are also shown.

Time (hours after exposure)	Hour of the day	Exchange velocity ($m\ s^{-1}$)	Flux* ($Bq\ s^{-1}\ m^{-2}$ soil)	Plant mass (kg fw m^{-2} soil)	LAI	Dry matter production rate (kg $CH_2O\ s^{-1}\ m^{-2}$ soil)
-1	10	7.86×10^{-4}	-2.24×10^{3}	3.59	4.70	6.19×10^{-9}
0.2	11	8.85×10^{-4}	1.37×10^{2}	3.59	4.70	1.64×10^{-8}
1	12	8.85×10^{-4}	1.31×10^{2}	3.59	4.70	1.64×10^{-8}
24	11	6.30×10^{-4}	4.85×10^{1}	3.57	4.60	-1.34×10^{-8}
120	11	6.63×10^{-4}	4.28	3.51	4.20	-6.12×10^{-9}
336	11	6.21×10^{-4}	2.26×10^{-3}	3.38	3.30	-6.84×10^{-9}
432	11	6.94×10^{-4}	3.08×10^{-2}	3.33	3.00	-2.98×10^{-9}

*from plant to air

6.2 HTO Concentrations (SB1 and SB4)

Leaves and Stems: The predicted HTO concentrations in leaves and stems were higher than the observations immediately after the exposure, by a factor of 4 for SB1 and a factor of 2 for SB4. This suggests that the model overestimates the HTO transfer rate from air to leaves. The degree of overestimation increased through hour 1 for SB1 and through hour 24 for SB4 but decreased thereafter, resulting in predictions that were lower than the observations by 120 hours in each run. This implies that losses of tritium from the leaves occur too rapidly in the model in the period 1-4 days following exposure. Nevertheless, the model performed well over the first 24 hours, the period of high concentration that determines the total amount of OBT formed in the leaves. The large underpredictions beyond 120 hours are believed to arise because ETMOD does not allow for the slow conversion of OBT to HTO.

Shells and Seeds: As noted above, the initial HTO concentration in the pods is assumed to be half the concentration in the leaves. This resulted in underpredictions for SB1, where the leaf concentrations were at background levels at the time the pods formed. For SB4, the assumption also resulted in an initial underestimation, by a factor of about 2, which suggests that the HTO taken up during the exposure moves rapidly through all parts of the plant. However, a biological half time of 2 days for the HTO in the pods appears to be too long, since the predictions rose above the observations beginning at 24 hours. The predictions dropped below the observations again at about 500 hours because ETMOD does not allow for the slow conversion of OBT to HTO. The model performed well over the first 24 hours when the concentrations were high.

6.3 OBT Concentrations

Leaves and Stems: ETMOD predictions for OBT concentrations in leaves and stems at harvest agreed well with the observations for exposures that took place before any pods had formed. However, for later exposures, ETMOD assumes that all OBT formed in the leaves is translocated to the pods and sets the leaf concentration to background levels. This assumption was not supported by the observations, which show that some OBT is retained in the leaves even when the exposure occurs at late growth stages, with the result that ETMOD severely underestimated these endpoints.

Shells and Seeds: ETMOD's predictions of OBT concentrations in the pods agreed well with the observations for exposures that occurred after the pods had formed (SB3, SB4, SB5 and SB6). In contrast, the model severely underpredicted the concentrations in pods that had not yet started to form at the time of exposure (SB1 and SB2). In this case, the predicted HTO concentrations in the leaves had dropped off to very low values by the time the pods had started to form, so that the dry matter translocated to the pods was essentially uncontaminated with tritium.

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Belot Model Description and Discussion of Results

1. MODEL DESCRIPTION

To evaluate the accumulation of tritium in the organic matter of plant organs such as fruits, grains, roots or tubers, our preliminary approach is the following. It is assumed for simplification that: (i) the growth rate of the organ is constant during the linear growth phase of the organ and negligible outside this phase; (ii) the organic matter formed in foliar tissues is transported to the growing organ by translocation. At each time, the specific activity of the newly formed organic products (expressed in activity of combustion water) is proportional to the specific activity of leaf water. The final specific activity of the organic matter in the organ at harvest is then proportional to the mean specific activity of leaf water during the whole linear growth phase of the organ in question (Belot, 1996). This is the basis of the following model, which was further refined to take into account the influence of variations in light.

The specific activity of the organic matter of a given storage organ at harvest C_{OBT} is thus calculated by determining a weighted mean activity of leaf water C_{HTO} over the duration T of the linear growth phase of the organ. This is expressed by:

$$C_{OBT} = \frac{\alpha}{T}\int_0^T C_{HTO}(t)g(t)dt \qquad (1)$$

where $\alpha = 0.6$ is a dimensionless fractionation ratio defined as the ratio of the specific activities of combustion water and tissue water in equilibrium conditions;
T is the duration of the linear growth phase, which is rather well documented for the most important crops; and,
$g(t)$ is a corrective weighting factor that expresses the influence of the diurnal light flux variations on carbon assimilation and therefore concomitant tritium incorporation.

The corrective weighting factor $g(t)$ was introduced in the model equation to take into account the influence of the light flux on the growth rate of the organ at a small time scale. If the growth rate is proportional to the light flux, this dimensionless factor should be equal to the ratio between the light flux at time t and the average light flux over the whole duration of the growth phase (including night). If a light saturation effect is expected, the real light flux should be replaced by the efficacious light flux, which, in a first approximation, can be set equal to the real flux when saturation does not still occur, or to the saturation flux otherwise.

2. APPLICATION TO THE SOYBEAN SCENARIO

Equation (1) is simplified by assuming, in a first approximation, that the totality of HTO absorbed in the leaves during exposure is flushed out of the leaves in an exponential way within a few hours after exposure and that practically no residual HTO remains in the leaves afterwards. If we suppose moreover that the exchange rate and light flux do not vary substantially during the phases of exposure and early exponential decline, we can integrate

Equation (1) and this yields the following much simpler equation:

$$C_{OBT} = C_{HTO}^{air} \; \alpha \; h \; g \; \frac{t_e}{T} \qquad (2)$$

where C_{HTO}^{air} is the concentration of tritium in air humidity during exposure;

h is the relative humidity of the atmosphere;

g is the corrective factor defined above as the ratio between the light flux during exposure and the mean light flux during the whole growth period;

t_e = 1 h is the duration of exposure;

T = 840 h (35 days) is the mean linear growth duration of the soybean seeds in normal conditions, as estimated from many references in the literature (*e.g.* Kumudini et al 2001, Petersen & Lauer 2004).

The simple model above allows to see that, under simplifying assumptions, the normalised concentration of OBT at harvest is directly proportional to the exposure duration and inversely proportional to the duration of the linear growth phase of the organ considered.

3. DISCUSSION OF RESULTS

The results obtained for the exposures SB4 to SB6 are given in Table 1. The seed growth period begins between SB3 and SB4, so that the experiments SB1, SB2 and SB3 cannot be treated by the simplified Equation (2). The most important parameter in the model is certainly the seeds linear growth duration T. This parameter does not represent the total duration of the growth phase, which is about 50 days, but the duration of the linear growth phase, which is generally estimated to be about T = 840 h (35 days). The latter value is the statistical mean of a great number of values for many plants in the field, different cultivars and different climatic conditions that may affect growth. The corrective factor g that characterizes the light influence at time of exposure is far from being negligible. This factor is calculated as explained above in model description, and amounts to 3.11, 2.44 and 2.16 for SB4, SB5 and SB6 respectively.

Table 1 : Results obtained by applying the simplified model to the proposed soybean scenario

Exp.	Observed C_{OBT} / C_{HTO}^{air} in pods and seeds at harvest	Predicted C_{OBT} / C_{HTO}^{air} in pods and seeds at harvest
SB1	8.63 E-06	*
SB2	2.44 E-05	*
SB3	5.28 E-04	*
SB4	2.61 E-03	1.46 E-03
SB5	1.56 E-03	1.37 E-03
SB6	0.40 E-03	1.24 E-03

*Cannot be estimated by using Equation (2), which assumes a sustained rapid exponential decrease of tritium in leaf water

For leaf exposure within the fruiting period (SB4 to SB6), the predicted concentrations in the seeds at harvest are close to the observed ones within a factor of about two. The somewhat greater difference for SB6 can be explained by the circumstance that exposure was carried out near the end of the linear growth period. Moreover, the simplifying assumptions of the model and the statistical nature of the input data can affect the three results obtained. Nevertheless, for the present scenario, the differences between the observed and predicted values are rather moderate, which comforts the tentative simple model presented above.

For pre-fruiting exposure (SB1 to SB3), the model does not provide any prediction, due to the assumption that the contamination of the seeds is quite negligible in this case. This does not correspond to the reality. In fact, some OBT is observed in the seeds, at a measurable concentration, which is nevertheless much smaller than observed for exposures during the fruiting period. This can be explained by the observation that the real time course of HTO–in–leaves after a short exposure is different from the time course assumed in the model. The elimination of HTO from the leaves is initially very fast as supposed for simplification, but becomes in fact slower and slower as time elapses. After a few days, there still remains in the leaves some amount of HTO that does not vary substantially throughout the growth period of the seeds and induces the accumulation of some OBT in the growing seeds. If we assume that the residual HTO in the leaves over the growth period of the seeds is comprised between 10^{-4} and 10^{-5} of the initial concentration of HTO in leaves, the normalised concentration of OBT in the seeds will be comprised in the same interval of magnitude. This agrees quite well with the observations made in the SB1 to SB3 experiments.

A prediction of OBT in seeds at harvest for pre-fruiting exposures could be obtained if it were possible to predict the real time course of HTO-in-leaves over a long time after exposure. Alternatively, the prediction could be based on the observed curve of HTO retention in soybean leaves or on a curve observed for other plants, assuming that it does not vary substantially with the plant considered. It seems that the form of the retention curve, while being governed in the beginning by the rapid turnover of water in leaves, is governed later on by the backward transport to the leaves, via the xylem path, of some of the HTO initially conveyed to the stem and roots via the phloem path. But, the residual HTO may also be due to a slight contamination of the soil water during the phase of leaf exposure, in spite of the precautions taken to avoid it. It seems that the first hypothesis is most probable, since the same form of HTO retention curve was already observed in much earlier experiments in which potato and vine leaves were exposed to HTO with precautions taken to avoid soil exposure (Guenot and Belot, 1984). Nevertheless, this needs to be substantiated by further observations, in experiments where the absence of soil contamination would be carefully verified by measuring HTO in soil samples at different times after end of leaf exposure.

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FzK Model Description

1. INTRODUCTION

The capabilities of the accident consequence assessment model UFOTRI were extended to consider a wider variety of foodstuffs. In a first step, rice was added to the list of foodstuffs and a generic rice model was developed. This type of model is used to perform the calculations for the soybean scenario. The newly developed model, however, is still not part of the UFOTRI distribution due to a lack of time for intensive testing.

2. MODEL DESCRIPTION

Basis of the modelling are processes that require light such as photosynthesis and photorespiration, and others which are independent on light, such as maintenance respiration and basic metabolism. Light-dependent and light-independent processes are treated in a different way. Light independent transfers were set to constant transfer rates whereas the light dependent transfer rates are described by physically based models. The reason behind this distinction is the lack of quantitative model approaches for the light independent processes, in particular for the basic metabolism.

Photosynthesis is calculated on the basis of net CO_2 assimilation by using an approach presented in Weir (1982) for wheat. However the approach can be adapted to other crops. This was successfully done in UFOTRI for vegetables, root crops and cereals. The derivation of the required parameter values together with factors taking into account several stress conditions can be found in Weir (1982) and Raskob (1993). Parameterisation specific to the new model is discussed below.

With the photosynthesis model it is possible to predict the build up and thus the growth of crops such as the rice plant and soybean. However what has to be adapted is the duration of the growth and the partitioning function. Before flowering, the whole organic matter production will end in the either stem, roots or leaves of the plant. After flowering, more and more material is directed towards the build-up of the seeds. Build up increases until, the linear growing phase starts, where the build up of material is nearly constant. This phase lasts several weeks followed by the drying of the seeds until maturity and harvest. The duration of the three phases can be selected by the user, dependent on the crop. The maximum photosynthesis rate was set to slightly higher values as for wheat.

The OBT incorporation into the edible part of the soybean per hour T_{act} is now directly related to the build up of organic matter and the concentration of tritium in the tissue free water

$$T_{act} = P_{act} * C_{TWT} * f_g * dis$$

where P_{act} is the actual hourly dry matter production rate in g/h,
C_{TWT} is the hourly mean TWT concentration in the crop in Bq/g,

f_g is a function describing the initial partitioning after flowering and before the linear growing phase, and
dis is a parameter taking into account that UFOTRI does only consider the whole plant and not the partitioning into leaves and stem (set to 2).

The function f_g is 1 during the linear growing phase. For the two other periods, f_g can be described as a sinusoidal function normalised to the duration of the phase. The duration of the three phases was assumed to be:
Phase 1: 30 days
Phase 2: 30 days
Phase 3: 30 days

A standard type of crop was applied for all calculations ignoring the variety of weights given in the scenario description. It was assumed that the overall uncertainty hides these variations in particular as the model is robust against these changes. Robustness means that when the crop weight is increased, also the build up of OBT is increased respectively. Only the initial specific HTO concentration in the crop may vary, however, such finesses might be considered in a second run.

3. PARAMETER VALUES

The parameters used for the soybean scenario are provided in Table 1.

Table 1. Parameter selection:

Parameter	value
Minimal stomata resistance	2 s/m
Plant water content at maximum	2000 g
Plant organic matter at maximum	500 g
Plant water content at maximum	100 g
Plant organic matter at maximum	600 g
Leaf area index at maximum	5 m^2/m^2
Constant concerning minimal PAR flux	30
Constant concerning water vapour deficit	0.2
Minimal temperature for stomata closure	8 °C
Maximal temperature for stomata closure	45 °C
Optimal temperature for stomata	28 °C
Day of harvest for rice	261
Time interval between anthesis and harvest	90 days
Duration of the first period after anthesis	30 days
Length of linear growing period	30 days
Length of maturity time before harvest	30 days

4. REFERENCES

Raskob, W. (1993). UFOTRI: Description of the New Version of the Tritium Model UFOTRI 4.0, including user guide. Report KfK-5194, Kernforschungszentrum Karlsruhe, 1993

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FSA Model Description

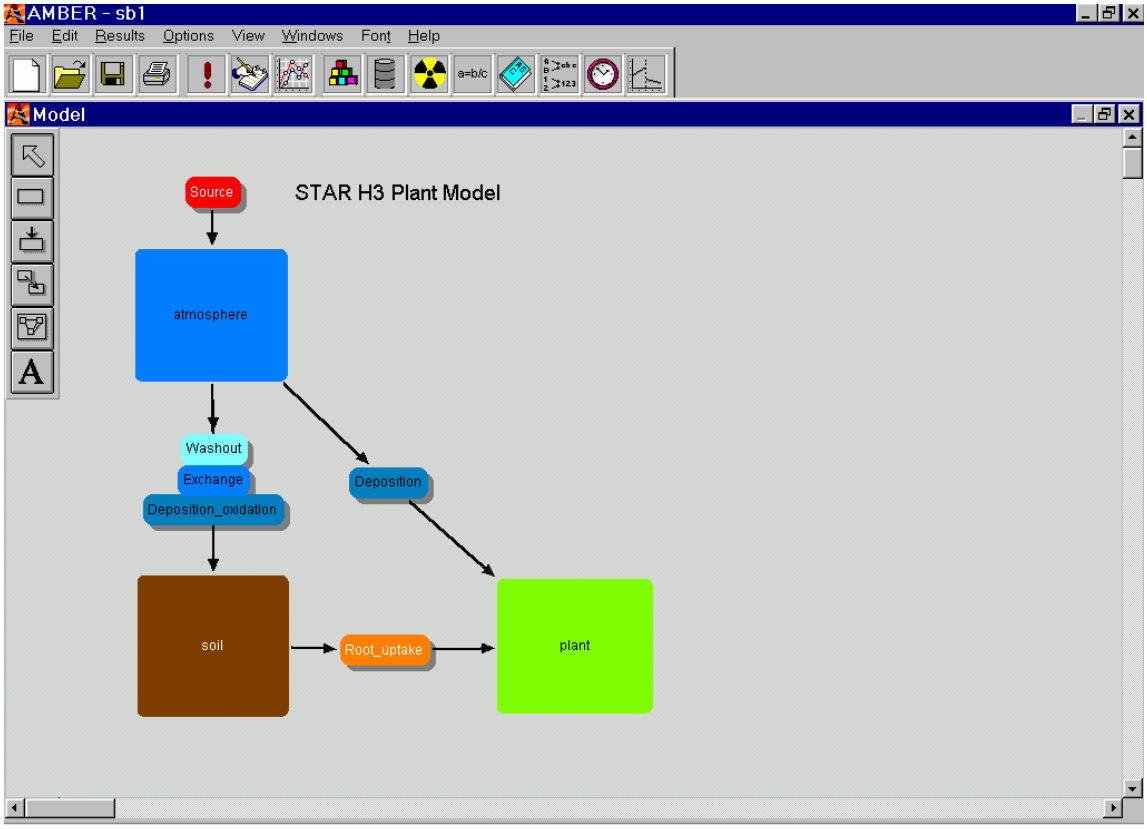
1. INTRODUCTION

Short-term discharges warrant special treatment as they may result in greater exposures to the critical group compared to the same activity discharged over a longer period. The reasons for this are two-fold. First, in the case of chronic discharges, these are assumed to be spread over a 360° wind rose over a year according to local weather patterns, whereas an acute release is usually released in a brief period within a small sector. This can result in higher concentrations particularly if the discharge is towards land and not sea. Second, over a short time scale, little weathering or nuclide decay will take place possibly resulting in higher concentrations in harvested crops and livestock. However, over a long time scale, concentrations in crops and livestock would decrease after an acute release.

2. MODEL APPROACH

The Food Standards Agency has developed the STAR-H3 model to determine the effect that short-term releases of H-3 have on the food chain. With ongoing development, the STAR H-3 model has now been incorporated into the compartmental model 'AMBER', which reproduces and enhances the behaviour of the original STAR-H3 model. These incorporate the methodology developed by Smith (1989) to take account of the short-term dynamic properties of these nuclides. This also incorporates the results of experimental work undertaken at Imperial College, London. The models include compartments that address losses from the plume through exchange with atmosphere, and through metabolic processes such as respiration. They also include compartments that allow for fixing of activity through photosynthesis in biota, translocation into storage organs in plants, and metabolism into carbohydrates, proteins and lipids in animals.

The following figure shows the basic outline of the compartmental model for STAR-H3 used in the soybean analysis.



2.1 Model compartments

Atmosphere. This is the concentration of H-3 in air surrounding the plant. This compartment is the source of H-3 for all other compartments. The model inputs for the compartment are the time integrated H-3 concentration in air, the water content in air and the time over which the concentration persists.

Soil. Soil in the root zone contains water and so hydrogen. Model inputs for this parameter are the bulk soil density and the soil water content. It is important to note that all of the tritium within this compartment is assumed to behave as HTO.

Plant (fast turnover). The proportion of the plant containing tritiated water. The model inputs for the compartment are the crop density, the areal evaporation rate and water content.

Plant (slow turnover). The proportion of the plant containing organically bound tritium, OBT. The model inputs for the compartment are the non-labile hydrogen content and mean residence time in the plant.

The two plant compartments need to be separately identified because of the different time constants for hydrogen retention and because OBT in compartment 4 has a higher value per unit intake.

3. TRANSFER FACTORS

There are 7 transfer factors that relate to the exchange of H-3 between compartments (in Figure 1) in the STAR-H3 plant model. It is a common occurrence that some foodstuffs may not have sufficient data to accurately model uptake. In such situations simple approximations are made concerning the genus of the plant.

- **Atmosphere to soil.** HT movement into soil and rapid oxidation to HTO, exchange of water between soil and atmosphere, wet deposition. For this scenario this transfer was set to zero to reflect the covering of the soil with polythene.
- **Soil to 'out of system'.** Losses due to exchange to atmosphere and loss to deep soil below the root zone.
- **Soil to plant 'fast'.** Uptake of water by the plant.
- **Plant to 'out of system'.** Evapotranspiration and exchange of HTO between plant and atmosphere.
- **Atmosphere to plant 'fast'.** HTO exchange, only HTO in the atmosphere
- **Plant 'slow' to Plant 'fast'.** Loss of tritium from non-labile or OBT.
- **Plant 'fast' to Plant 'slow'.** Rate of conversion of plant tissue water and other labile tritium to the non-labile form.

For all compartments except atmosphere an additional transfer is used to account for radioactive decay.

4. REFERENCES

Smith A.D. (1989) Calculation Of Critical Group Doses Resulting From Short-term Aerial Releases Of Radionuclides From Sellafield. BNFL, Risley.

GE Healthcare Model Description

1. MODEL DESCRIPTION

The GE Healthcare model is a dynamic compartment model formulated in terms of a series of coupled first-order differential equations. 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.

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.

2. PARAMETER VALUES

2.1 Generic parameter values

The generic parameter values within the model are provided in Table 1.

Table 1. Generic parameter values

Parameter	Value
Volume of the box	1.17325 m^3
Exchange velocity	0.0102 ms^{-1}
Plant fast turnover rate	1 h^{-1}
Residence time	32 days
g hydrogen per g water	1/9
Air turnover	0 during exposure, 1000 at all other times

3.2 Scenario specific parameter values

Parameter values specific to each scenario considered are provided in Table 2.

Table 2. Specific parameter values

Parameter	SB1	SB2	SB3	SB4	SB5	SB6
Activity concentration in the box (Bq m^{-3})	1.04E+12	1.97E+12	1.52E+12	7.02E+11	1.23E+12	1.83E+12
g water per kg plant [Plant fast water]	712	638.55	697.14	680.44	672.5	590.31
Water content of air (g water per m^3 air)	39.52	29.07	40.23	52.35	35.98	26.7

IFIN-HH Model Description and Discussion of Results

1. MODEL DESCRIPTION

An improved version of the tritium module in the EC project RODOS was used, initially developed in our institute in collaboration with FZK-Germany (Galeriu et al, 2000). The model has a similar general structure as UFOTRI but the transfer parameters for tritium are derived from plant physiology. The transfer of HTO from air to leaves is modeled with an exchange velocity that includes a canopy resistance. The canopy resistance is modeled using a physiological model depending on canopy photosynthesis rate. The leaf conductance to CO_2 is given by,

$$g_{sc} = g_c + \frac{A_g \cdot (0.9 - \frac{4.7}{A_{m,g}} \cdot \frac{D_s}{D_{\max}})}{C_s - C_{i,vir}} \quad C_i = f(C_S - \Gamma) + \Gamma \quad f = f_o(1.-D_S / D_{\max})$$

where C_s and C_i are the CO_2 concentration at leaf surface and leaf interior,
Γ is the CO_2 compensation point,
D_s is the humidity deficit at leaf surfaces,
f_o and D_{max} are parameters describing the effect of humidity deficit on leaf resistance,
g_{sc} is the stomata conductivity for CO_2,
g_c is the cuticle conductivity,
A_n and A_g are the net and gross photosynthetic rate,
A_{min} is the residual cuticle photosynthetic rate,
$A_{m,g}$ is the maximum gross photosynthetic rate, and,
Rd is the dark respiration rate.

By integrating over the canopy, the canopy conductance is directly linked to the canopy photosynthesis. For the last quantity we use the submodel from the crop growth model WOFOST with accompanying physiological plant parameters in the database (Melintescu et al, 2002). Note that the dry biomass considered is obtained after extraction of maintenance and growth respiration while the canopy resistance use the initial photosynthetic rate.

The conversion of HTO to OBT is driven by photosynthesis rate, using stoichiometry and an isotopic discrimination rate. We consider the net OBT formation, after maintenance and growth respiration processes. Part of the newly formed OBT is distributed to grain (pod), in accordance with dry matter partition (plant and cultivar specific). The model includes also OBT formation in night, but this is not relevant in the present scenario.

2. ADAPTATION TO THE SOYBEAN SCENARIO

In order to model the biomass dynamics for the present scenario and soybean cultivar, we have constructed a daily weather file, combining scenario data with climatic ones (monthly mean values from Wolsong area in INTERNET were used to generate a daily sequence using free software (wgen.for). An hourly meteorological file was constructed by interpolation from

scenario data and further used in the tritium model. We start with an existing tropical cultivar in the database and have slightly adapted the parameters in order to obtain the dynamics of biomass growth as given in the scenario. In our model, the plant growth is modeled in a simplified way but still gives reasonable results. We note that the scenario data have a large variability of biomass at harvest between experiments, a coefficient of variation of 60 % for seeds and 30 % for total biomass. These can influence the model prediction uncertainty. There are no direct data on leaf area index dynamics. These shortcomings of the input data give some uncertainty in the plant biomass and LAI dynamics. Two plant models were considered, one with minimum biomass and LAI and one with maximum one.

The biomass dynamics from experiment and the two variants of the soybean model are given in Figure 1. Seed mass at harvest was most accurately predicted by model 1 and total biomass by model 2.

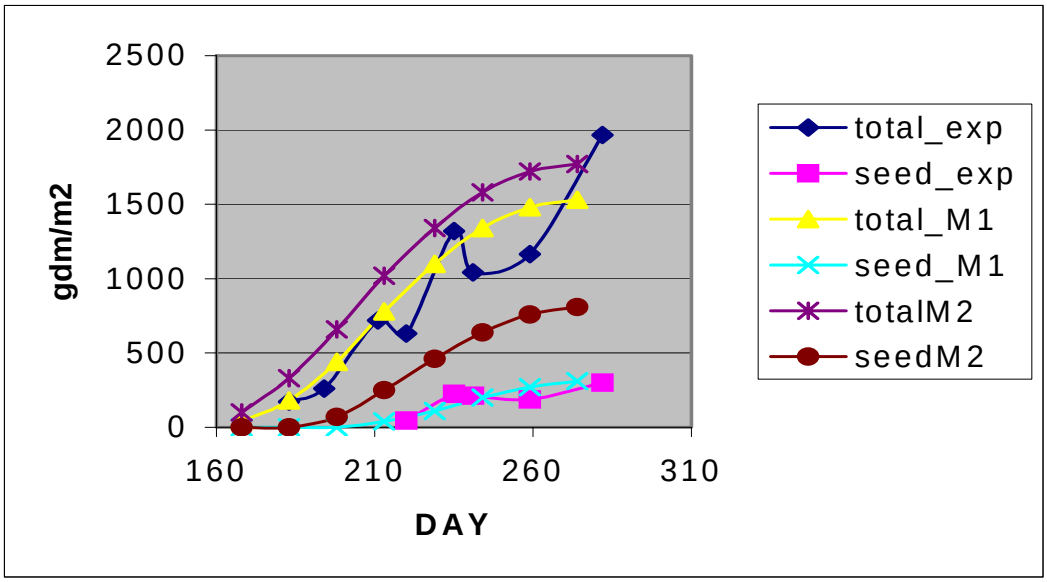


Figure 1. Biomass dynamics: experiment and models.

Figure 2 gives the model 1 LAI dynamics in comparison with the prediction of plant growth model WOFOST. We note that for SB1, the predictions are lower than the best estimate of LAI. Indeed, from the leaves dry mass and the specific leaf area of soybean (derived from the literature) we deduced a LAI of 3 (at this stage all leaves are green), while model 1 predicted a value of 1.35. Model 2, with increased biomass, gave the right LAI at SB1, but higher LAI later.

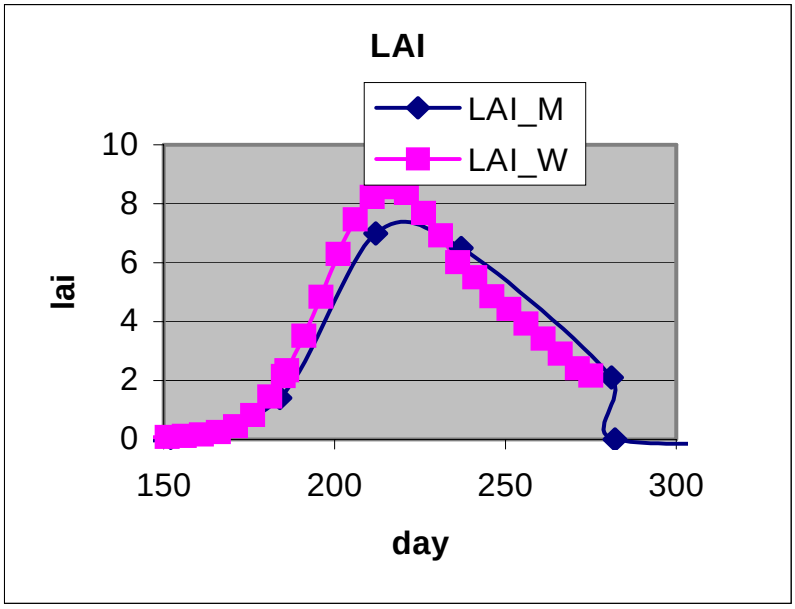


Figure 2. LAI in tritium model and WOFOST model and RODOS-H result

Among intermediary model results we present the predictions for the canopy resistance as functions of time and experiment in Figures 3 and 4.

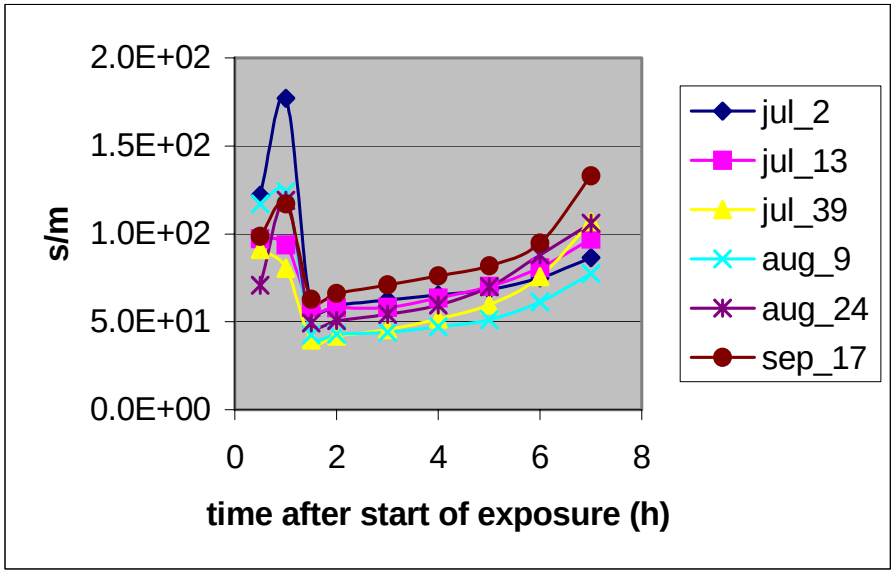


Figure 3. Canopy resistances in the first 8 hours from the start of the exposure

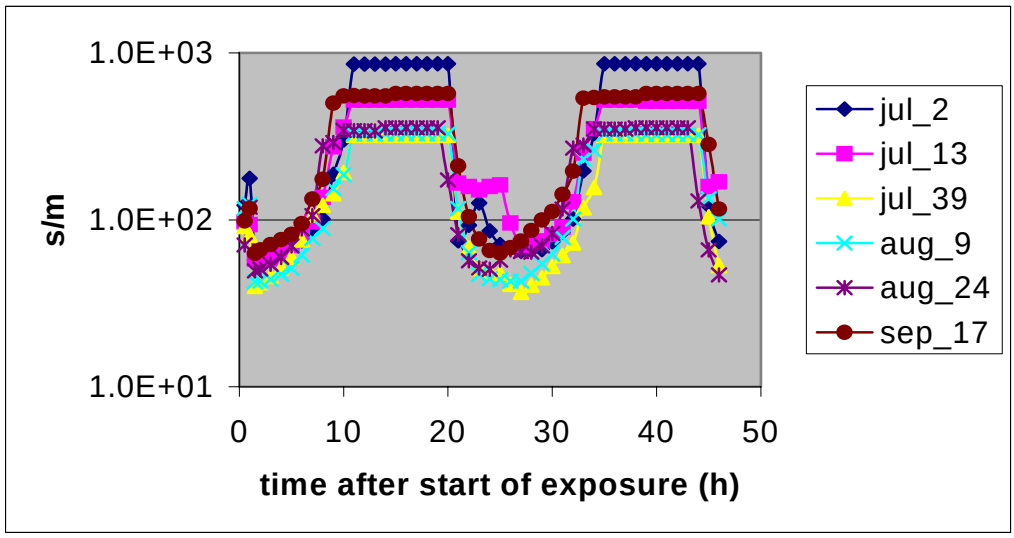


Figure 4. Canopy resistance for first 46 hours after start experiment

The largest canopy resistance in the light is for the experiment in July 2, in the period of fumigation but also after. The higher resistances in the periods of box experiments can be a result of more factors:

- Increased temperature in the box, as for Aug 9 where we have values of 45-48 °C- definitely depressing photosynthesis and increasing the canopy resistance.
- Under-prediction of solar radiation. A general conversion factor of 5.5 was used to convert luminance to solar radiation. This gives value of solar radiation in the box much lower than that measured (up to a factor 2).

Higher resistance occurs for the first experiment but also for the last (Figure 4) as a result of plant phenology and development. In SB1, the plant was young with few leaves but all green. In SB6, the plant is old with few green leaves.

The results presented partially explain the large difference in the dynamics of HTO concentration of leaf water between experiments, as seen in Figure 5.

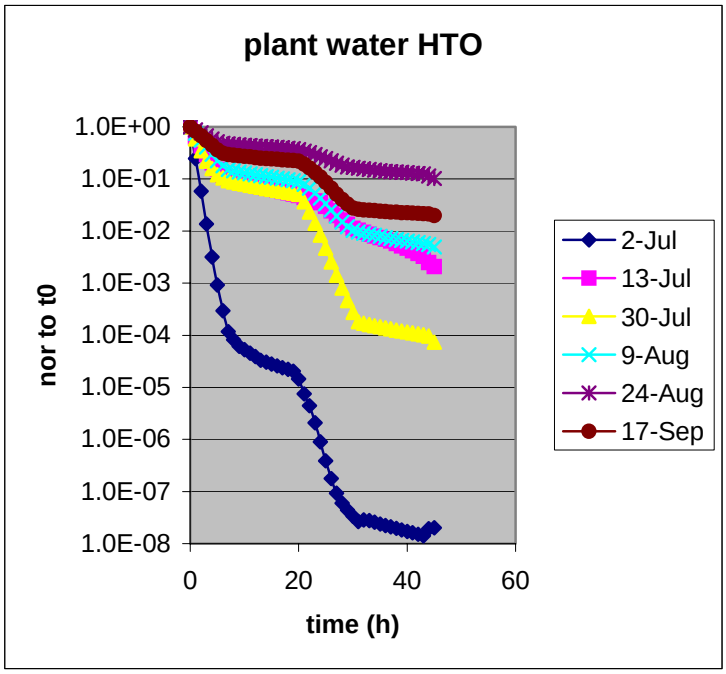


Figure 5. Leaf water HTO concentrations normalized to end of fumigation

OBT concentration in seeds depends on daily OBT production and on the partition to storage organs. No information on the specific Korean cultivar used in the experiments was available and literature values are variable. OBT production is dependant on leaf water HTO concentration and photosynthesis rate.

The growth period is divided into vegetative and reproductive periods. Emergence is considered at development stage 0, flowering at development stage 1 and harvest at development stage 2. The partition to storage organs as in WOFOST database for soybean was initially used.

No data on the translocation to storage organs before flowering was available, but it is known that a part of the new dry matter stored in stems at flowering stage can be potentially translocated to grain (pod) and this fraction is plant dependant (Table 1, Penning de Vries, 1989).

Table 1. Fraction of stem weight at flowering potentially translocatable to storage

Plant	Fraction
Soybean	0.18
Wheat	0.4
Faba bean	0.45
Potato	0.3

In absence of information on the Korean cultivar an intermediate situation was considered with low translocation during the flowering stage (fraction of stem weight subject to translocation between 0.05-0.15).

3. DISCUSSION

The model was variable in its predictions of HTO and OBT in relation to the observed values. The HTO concentration in leaves was over-predicted by a factor of 3 to 5 in experiments SB1 and SB4 at the end of exposure, but under-predicted by 40 - 90 times at harvest. OBT in pods was under-predicted by 100 times in SB1, but over-predicted by 10 times in SB6.

A number of sources of uncertainty were analysed:

- Wind speed – unknown anemometer height. This may have a marginal effect on atmospheric resistance and exchange velocity.
- Improper plant LAI and biomass in the model, compared with data used. This could be a potential source of error as the simple growth model used is not appropriate for measured biomass and there is uncertainty in the scenario LAI. This can explain our fast release in SB1, due to under-prediction of leaf biomass and LAI.
- Variability of experimental harvest biomass among experiments. There is a large spread of experimental data and a proper growth trend is therefore difficult to assess. At harvest the total dry biomass is on average 1966 g/m^2, but with a range of 1286-3225. A factor 2 misprediction in the canopy resistance and OBT production from the variability of biomass production between experiments can be expected on this basis.
- Ambiguities in the scenario relating to the large water content in seed at harvest. Based on general agricultural practice, a water content no more than 20 % in seeds at harvest would be expected. However, in the scenario data the water content is close to 60%.
- Difficulties in assessing the proper characteristics of the Korean cultivar. Such as translocation from stem to grain, grain filling dynamics, temperature effect on photosynthesis.
- The covering applied to the soil during the exposure may not have been completely effective at preventing a small amount of tritium from depositing to the soil. Root uptake from the soil may then have acted to keep the HTO concentrations in the plant at a relatively high level.

The general over-prediction in HTO at the end of exposure period may result from the unusually high temperature in the exposure chamber. The average temperature for SB1 - SB6 was 40, 33, 39, 47, 40 and 32 °C, respectively. The maximum environmental temperature was only 34 °C. Plants cease photosynthesis at high temperature, the cut-off value depending on plant type and the adaptation to average environmental conditions. We have no idea on the temperature cut-off for the Korean cultivar but literature values are lower than 47 ° C. Penning de Vries (1989) gives the following values (Table 2).

Table 2. Temperature dependence of maximum photosynthesis rate

Temperature (°C)	Factor
0	.0001
10	.3
20	.6
25	.8
30	1
35	1
40	.8
50	.001

It seems highly probable that in SB4 there is a depression of uptake and photosynthesis while in SB2 and SB6 this is excluded. Both SB1 and SB4 show a low uptake, Cplant/Cair is around 0.125, which implies an uptake rate near 0.33 h^{-1}. Therefore, it appears that plants were under stress in the chamber in SB1 and SB4 and this was not taken into account by most of the modellers.

4. REFERENCES

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F W T Penning de Vries, D M Jensen, H F M ten Berge and A Bakema, 1989, Simulation of ecophysiological processes of growth in several annual crops Pudoc Wageningen.

Japanet Model Description

1. JAPANET MEMBERS LIST

NIRS (Kiriko Miyamoto, Yoshikazu Inoue, Hiroshi Takeda, Kazuhide Yamamoto)
Ibaraki University (Michiko Ichimasa, Yusuke Ichimasa)
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2. ASSUMPTION FOR CALCULATION OF SOYBEAN SCENARIO

1. Model and parameters are mostly based on the observation in Ibaraki University's semi-field release experiments in 1999-2002 of deuterium oxide vapor exposed to soybean (Table 1, Ichimasa et al., 2002, 2003).
2. No difference of TFWT in each part of a soybean plant. Errors of TFWT concentration were estimated from 10% variation of the rate constant of HTO loss from plant in the model.
3. Accumulation rate of non-exchangeable OBT (nOBT) in seeds changes depending on growing stages of soybean plant. Errors of nOBT concentration were estimated from the standard deviations of mean HTO concentration in air vapor during the exposure.
4. No consideration of soil properties, biomass balance of plant, meteorological and artificial conditions in the glove box and in the field.

Table1. Comparison of characteristics of soybean scenario and Ibaraki University's experiment

	Soybean scenario	Ibaraki University's experiment
Tracer	HTO vapor	HDO vapor
Release Time Duration	1 hour	8 hours
Exposure Conditions	20-50^{o}C, 50-90% humidity	20-30^{o}C, 50-90% humidity
TFWT Measurements During Exposure	No	Often to observe a rate constant of D_2O uptake from air
TFWT Measurements Just After Taking Out	3 times	Often to observe a rate constant of D_2O loss from plant
TFWT Measurements Until Harvest	5 times	No
nOBT Measurements After Taking Out	At final yellow bean harvest	Often until final yellow bean harvest including the stage of green bean harvest

3. Ibaraki University's model

Belot's equation (Belot et al, 1979) was modified:

1. Uptake of deuterium by the soybean plant is expressed as:

$$Crp = Ca \times Crmax \times [1-\exp(-k_1 t)]$$

where

Crp = tissue free water deuterium (TFWD) concentration ratio in plant (ppm)
Ca = deuterium concentration in air moisture around sampling point at time t (ppm)
Crmax = steady-state concentration ratio (Crp/Ca)
k_1 = rate constant of D_2O uptake from air (h^{-1})
t = time after the start of exposure (h)

2. Loss of deuterium from the soybean plant is expressed as:

$$Cp = C_0 \times \exp(-k_2 t)$$

where

Cp = TFWD concentration in plant (ppm)
C_0 = TFWD concentration in plant at time t=0 (the end of release) (ppm)
k_2 = rate constant of D_2O loss from plant (h^{-1})
t = time after the end of release (h)

3. OBD translocation to bean from leaf is expressed as:

$$TLIa = OBD/Ca$$

where

TLIa = translocation index of OBD to bean (%)
OBD = OBD concentration in bean at harvest (ppm)
Ca = mean D_2O concentration in air moisture at steady state (ppm)

4. REFERENCES

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KAERI Model Description

1. INTRODUCTION

Tritium (as HTO) released from nuclear facilities is readily absorbed to plants by photosynthesis, and changes into a constituent tritium of organic compounds by metabolism. The organically bound tritium (OBT) is generally non-exchangeable and remains in tissue of plant after the time of harvesting so that it can be an important contributor to dose (Barry et al., 1999). To assess potential does to human from accidental releases, it is necessary to model the behavior of tritium in the environment. To this end a number of dynamic models have been developed and their capabilities have evaluated and compared through the international studies (BIOMOVES II, 1996a and 1996b; IAEA 2001).

In 2003, International Atomic Energy Agency (IAEA) started on a new international joint research program, EMRAS (Environmental Modeling for Radiation Safety) succeeding BIOMASS program (IAEA, 2001). The EMRAS was organized to test the accuracy of model predictions and to improve existing models and specify their parameters. This paper describes the model prediction for the scenario of tritium absorption by soybean foliage submitted to Tritium-Working-Group of EMRAS (Theme 1, task 2).

2. MODEL DESCRIPTION

For model prediction, a dynamic compartment model (ECOREA-GH3) that was developed by KAERI (Korea Atomic Energy Research Institute) on the basis of the long-term model of UFOTRI (Raskob, 1990,1993) was used. The model was specially designed for evaluating the transfer of tritium into the grain-plant growing in dry-fields such as wheat and soybean after an acute release from a nuclear facility. Figure 1 shows the compartments and transfer pathways of the model. The plant is divided into four compartments: HTO and OBT compartments of plant body (stem + leaves), and HTO and OBT compartments of grain, respectively. The soil is divided into three compartments: layers of 0-5 cm, 5-15 cm and 15-30 cm. There is a reversible tritium exchange between all the plant compartments except the OBT compartment of grain in which all the organically bound tritium is insoluble and remains there after the time of harvesting. Water absorption of plant from soil occurs all via only the body of plant.

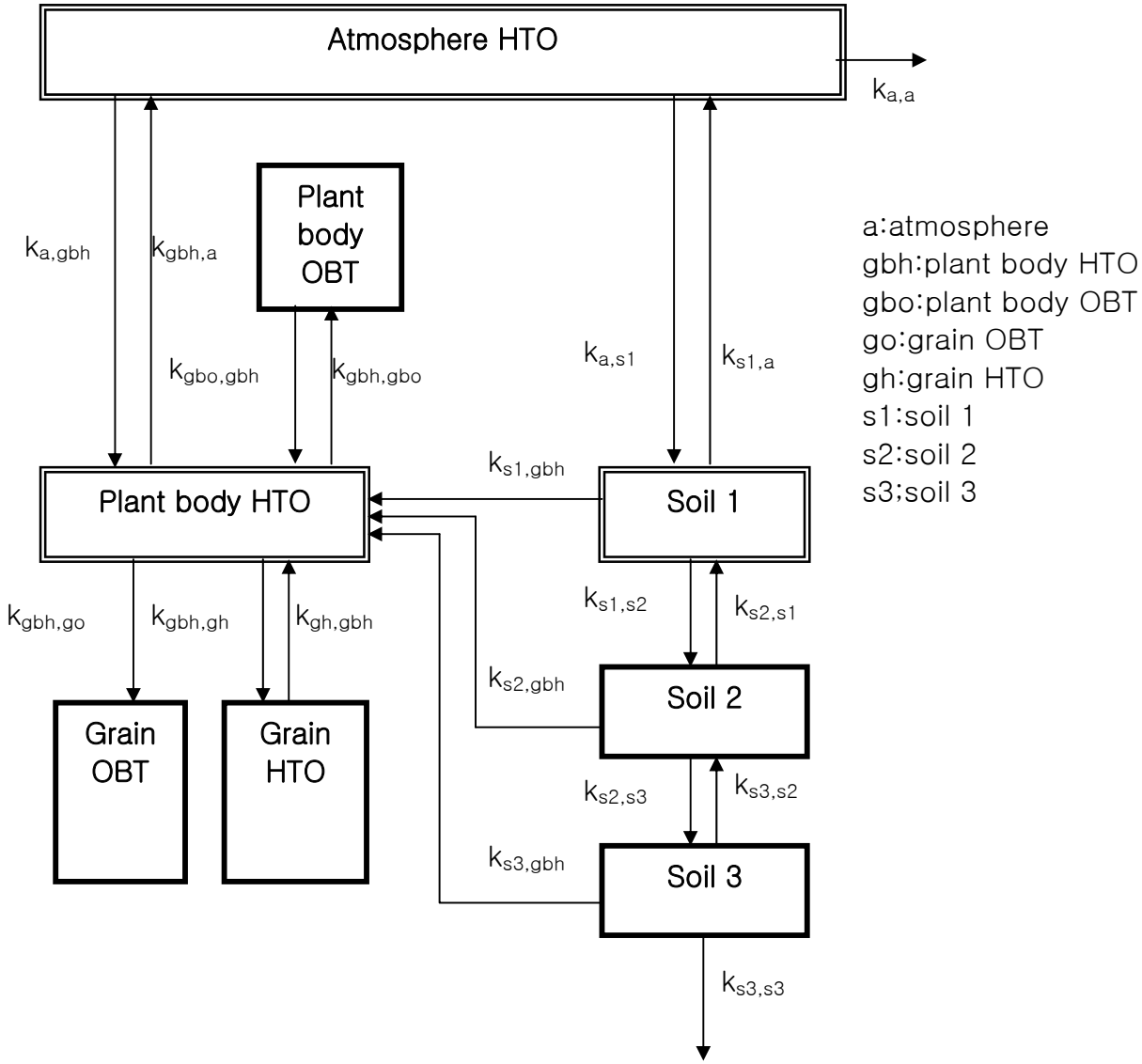

Figure 1. Compartments and transport pathways for ECOREA-GH3

The mass transfer between compartments can be generally described as

$$\frac{dA_i}{dt} = \sum_{k=1}^{m} K_{k,i} A_k - \sum_{i=1}^{n} (K_{i,j} + \lambda) A_i \qquad (1)$$

where

A_i (Bq/m^2) is the activity of compartment i,

$K_{k,i}$ is the transfer rate from compartment k to i, and,

λ is the decay constant of tritium (6.44×10^{-6} h^{-1}).

2.1 Biomass equation. The hydrogen inventory of plant varies with the growth of biomass, and it subsequently influences the transfer rate between compartments. Figure 2 shows the growth curves of soybean obtained with the biomass data presented in scenario. All data were fitted to the typical sigmoid growth curve with three parameters.

$$B(t) = \frac{B_1 B_2}{(B_1 - B_2) e^{-B_3 t} + B_2} \qquad (2)$$

The parameters are summarized in Table 1. The difference of the weight between the dry and fresh biomass at time t is assumed to be equivalent to the weight of water of the HTO compartment of plant.

2.2 HTO deposited during exposure. During exposure, the amount of HTO deposited onto the soybean plant was calculated using the Belot equation (Belot, 1979; Amano and Garten, 1991).

$$C_{gbh}{}^{o} = \alpha \times R_{ini} \times C_a{}^{o} (1 - e^{-\tau \Delta t}) \qquad (3)$$

where,

$C_{gbh}{}^{o}$: tritium concentration in body tissue water, Bq/kg
R_{ini} : mean relative humidity of air during exposure
$C_a{}^{o}$: mean activity of tritium in air moisture during exposure, Bq/kg
τ : time constant until equilibrium which is defined by $\rho_{s,ini}/(\alpha \mu_{ini} \gamma_t)$, h^{-1}
$\rho_{s,ini}$: saturated air humidity during exposure, kg/m^3
μ_{ini} : water content of plant body at the time of exposure, kg/m^2
α : H/T isotope ratio in air and plant (1.1)
γ_t : total resistance from atmosphere to stomata, h m^{-1}, and,
Δt : exposure time, h

At equilibrium,

$$C_{gbh}{}^{o} = \alpha \times R_{ini} \times C_a{}^{o} \qquad (4)$$

On the other hand, there was no the tritium deposited onto soil because it was covered with a vinyl paper during the exposure to the plant.

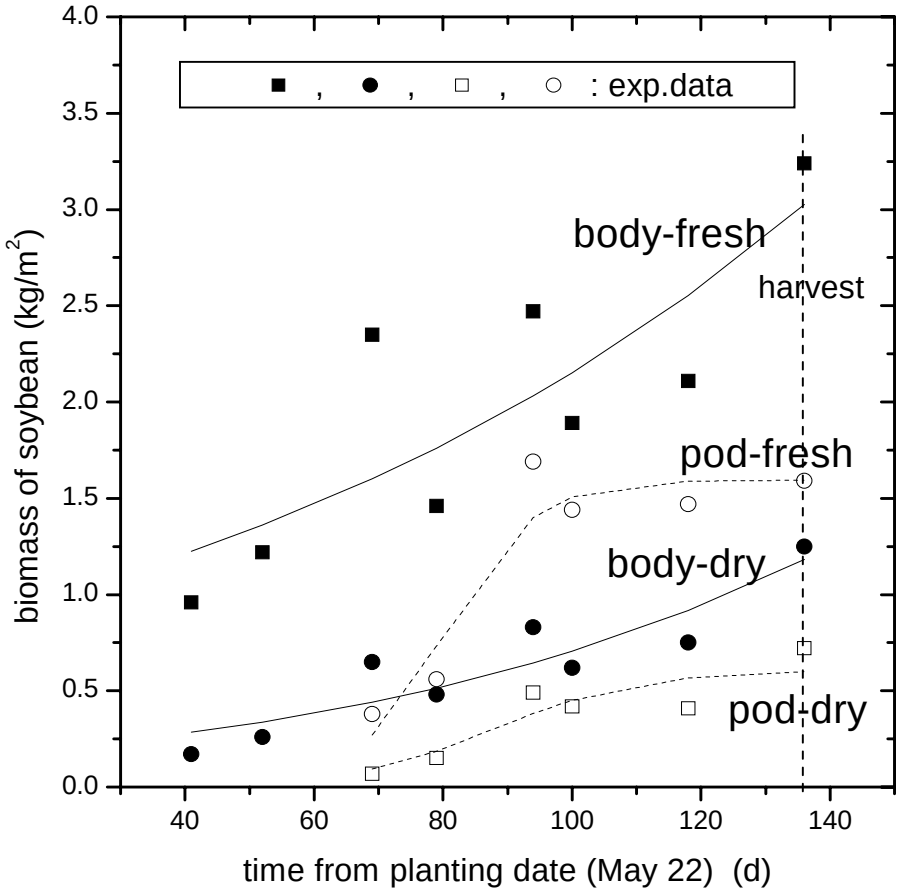

Figure 2. The growth curve of soybean

Table 1. Parameter values for the growth curve of soybean

parameter / plant	B1 (kg/m2)	B2 (kg/m2)	B3 (d)
Body (fresh)	1.87E2	0.827	9.6E-3
Body (dry)	7.10	0.147	0.017
Grain (fresh)	1.59	1.72E-5	0.143
Grain (dry)	0.61	2.43E-4	0.089

3. INPUT DATA

3.1 Basic data.

In order to calculate the transfer rate between compartments in the model, the following basic input data were used (Table 2). Some of the data came from the UFOTRI (Raskob, 1990), and others from the experimental condition presented in scenario.

Table 2. General input data

Parameter	Value
Mean height of the air mixing layer (H_m)	1000 m
Mean deposition velocity of HTO to soil (V_d)	18.0 m/h
Water humidity of saturated air at 25°C (ρ_s)	0.024 kg/m^3
Mean relative humidity during the growth of soybean (RH_a)	84%
Mean rainfall rate during the growth of soybean (K_{rain})	1.0 kg/(m^2.h)
Thickness of soil layer 1 (d_1)	0.05 m
Thickness of soil layer 2 (d_2)	0.1 m
Thickness of soil layer 3 (d_3)	0.15 m
Mean moisture content in soil (θ)	0.2
Fraction of root uptake of water from soil layer 1 (F_1)	0.2
Fraction of root uptake of water from soil layer 2 (F_2)	0.4
Fraction of root uptake of water from soil layer 3 (F_3)	0.4
Water content of plant body ($\mu(t)$)	B_{body}(fresh)-B_{body}(dry)
Activity ratio between the plant water and the water vapor at equilibrium (R_a)	0.5
Growing period of bean (T_{gg})	1440 h (60 days)
Half-time of tritium loss from plant body OBT (T_{gbo})	240 h (10 days)
Half-time of tritium loss from plant body HTO (T_{qbh})	2 h

3.1 Transfer rate.

The transfer rates are calculated on the basis of hydrogen inventory and hydrogen exchange between compartments with the assumption of equilibrium. The hydrogen inventory (kg/m^2) of each compartment is calculated by,

$M_a = H_m \times \rho_s \times RH_a \times 11\%$ for atmosphere compartment
$M_{s1} = 1000 \times d_1 \times \theta \times 11\%$ for soil 1 compartment
$M_{s2} = 1000 \times d_2 \times \theta \times 11\%$ for soil 2 compartment
$M_{s3} = 1000 \times d_3 \times \theta \times 11\%$ for soil 3 compartment
$M_{gbh} = (B_{body}(\text{fresh}) - B_{body}(\text{dry})) \times 11\%$ for body HTO compartment
$M_{gbo} = B_{body}(\text{dry}) \times 8\%$ for body OBT compartment
$M_{gh} = (B_{grain}(\text{fresh}) - B_{grain}(\text{dry})) \times 11\%$ for grain HTO compartment
$M_{go} = B_{grain}(\text{dry}) \times 8\%$ for grain OBT compartment

The hydrogen content in organic part of plant was assumed to be 8%. Transfer rate for the system is summarized in Table 3. The transfer rate of loss of HTO from atmosphere ($K_{a,a}$) was determined with the assumption of the half-time of loss of one hour, but the value of $K_{a,a}$ of 100 was assumed for the time less than 0.1hr in order to consider the effect of ventilation by an external fan just after the exposure. The rate constant of loss of HTO from plant during day-time ($K_{gbh,a}$) was assumed to be inversely proportional to the water content (μ) of the plant, with the reference value of 0.347 that is equivalent to the half-time of loss of one hour when μ is 0.4 kg/m^2. Since the water content of plant varies with the growth of biomass, the rate constant $K_{gbh,a}$ is time-dependent.

Table 3. Transfer rate between compartments

Transfer rate	from	to	Value in h^{-1}
$K_{a,a}$	Atmosphere	Outside	0.693 for t>0.1 hr, 100.0 for t<0.1hr)
$K_{s3,s3}$	Soil 3	Deep soil	3.42×10^{-4} (Raskob, 1990)
$K_{gbh,a}$	Body HTO	Atmosphere	$0.139/\mu(t)^*$, 0.347 for μ=0.4 kg/m^2
$K_{a,gbh}$	Atmosphere	Body HTO	$R_a K_{gbh,,a} M_{gbh}/M_a$
$K_{a,s1}$	Atmosphere	Soil 1	$V_d/H_m + K_{rain} / M_a \times 11\%$
$K_{s1,a}$	Soil 1	Atmosphere	$(K_{a,s1}M_a - K_{s3,s3}M_{s3} - (1-R_a)/R_a K_{a,gbh} M_a)/M_{s1}$
$K_{s2,s1}$	Soil 2	Soil 1	$K_{s3,s3}M_{s3}/M_{s2}$
$K_{s3,s2}$	Soil 3	Soil 2	$K_{s3,s3}M_{s3}/M_{s3}$
$K_{s1,gbh}$	Soil 1	Body HTO	$(1-R_a)/R_a K_{a,gbh} M_a/M_{s1}F_1$
$K_{s2,gbh}$	Soil 2	Body HTO	$(1-R_a)/R_a K_{,agbh} M_a/M_{s2}F_2$
$K_{s3,gbh}$	Soil 3	Body HTO	$(1-R_a)/R_a K_{a,gbh} M_a/M_{s3}F_3$
$K_{s1,s2}$	Soil 1	Soil 2	$(K_{a,s1}M_a + K_{s2,s1}M_{s2})/M_{s1} - (K_{s1,gbh} + K_{s1,a})$
$K_{s2,s3}$	Soil 2	Soil 3	$(K_{s1,s2}M_{s1} + K_{s3,s2}M_{s2})/M_{s2} - (K_{s2,s1} + K_{s2,gbh})$
$K_{gbo,gbh}$	Body HTO	Body OBT	$0.693/T_{gbo}$
$K_{gbh,gbo}$	Body OBT	Body HTO	$K_{gbo,gbh}M_{gbo}/M_{gbh}$
$K_{gbh,gh}$	Body HTO	Grain HTO	$0.693/T_{gbh}$
$K_{gh,gbh}$	Grain HTO	Body HTO	$K_{gbh,gh}M_{gbh}/M_{gh}$
$K_{gbh,go}$	Body HTO	Grain OBT	$1.386 \times M_{qo}/(T_{qq}M_{qbh})$

* $\mu(t)=B_{body}(\text{fresh})-B_{body}(\text{dry})$

4. RESULTS

Modelers were asked to calculate:
(1) TWFT (tissue-free-water-tritium) concentration of the body and pods for the SB1 experiment at the times: 0.2 hr, 1 hr, 24 hrs, 120 hrs, 336 hrs, 936 hrs, 1608 hrs, and 2280 hrs for body (stem and leaves), and 936 hrs, 1608 hrs, and 2280 hrs for pods (shell and seeds)
(2) TFWT concentration of the body and pods for the SB4 experiment at the times: 0.2 hr, 1 hr, 24 hrs, 120 hrs, 336 hrs, 768 hrs, and 1368 hrs for both body (stem and leaves), and pods (shell and seeds).
(3) The non-exchangeable OBT concentration of plant body and shell and seeds at harvest for the six experiments SB1 to SB6
(4) Estimate the 95% confidence intervals for all the predictions.

The results calculated for the questions (1) to (3) are given in Tables 4 and 5. All calculation results were obtained with the assumption that the HTO exchange between atmosphere and the tissue water of body during exposure was at equilibrium. This means that the initial condition of the body was determined by Eq.(4).

Table 4. Calculated TFWT concentration of body and pods with time for SB1 and SB4 experiment

SB1			SB4		
Time (hrs)	TFWT concentration of body (Bq/mL)	TFWT concentration of pods (Bq/mL)	Time (hrs)	TFWT concentration of body (Bq/mL)	TFWT concentration of pods (Bq/mL)
0.2	72000	-	0.2	23000	3300
1.0	64000	-	1.0	17000	11000
24	2200	-	24	2700	3000
120	8.2	-	120	9.2	9.7
336	4.6	-	336	3.1	3.1
936	1.2	1.2	768	1.1	1.1
1608	0.39	0.39	1368	0.31	0.31
2280	0.14	0.14			

Table 5. Calculated OBT concentration of body and pods at harvest for SB1 to SB6 experiments

case	OBT concentration of body at harvest (Bq/mL equivalent water)[1)]	OBT concentration of pods harvest (Bq/mL equivalent water)[1)]
SB1	0.84	0.07
SB2	3.65	2.38
SB3	9.5	127.7
SB4	7.4	86.0
SB5	48.4	320.7
SB6	450.1	592.8

1) One gram of dry matter is equivalent to 0.6 mL of combustion water

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LLNL Model Description and Discussion of Results

1. INTRODUCTION

Predictions for the Soybean Scenario were the result of manipulating output from the stochastic STAR-H3 model in Excel to account for processes missing in STAR and then using the Crystal Ball software to account for pathways to uncertainty missing in STAR.

2. MODEL DESCRIPTION

Primary modeling was done using STAR-H3, developed by QuantiSci. STAR is a compartmental model with inter-compartment transfer equations governed by user-defined parameters. Rates of transfers between compartments should be controlled by adjusting the parameters and not by altering the transfer rate equations. It's a very conceptually simple time-dependent model that, if run to equilibrium, maintains the T/H ratio from the air in the TFWT and somewhat increases it in the OBT. Although STAR accounts for different uptake and loss-rates of HTO between day and night, it does not account for plant growth or for changes in light-levels after exposure. Time-steps are hourly. STAR may be run either deterministically or stochastically.

There are four compartments (atmosphere, soil, tissue-free-water tritium (TFWT) in plants, and organically bound tritium (OBT) in plants).

HTO is deposited from atmosphere to soil through exchange with units of m^3 / (h kg); I zeroed the deposition velocity in this transfer, so the net deposition to soil was zero. HTO is also deposited via wet deposition, but of course that wasn't a pathway in this scenario.

HTO is deposited from atmosphere to plants:

$$Bq/m^3 \text{ x water content of plant (g } H_2O/\text{kg fw) / water content of air (g/m}^3) = Bq/\text{kg plant fresh weight (fw).}$$

There is a transfer from soil water to TFWT that uses evapotranspiration (g $H_2O/m^2/h$), crop density (kg fw/m^2 crop) and water content of soil (g H_20/kg), but the actual transfer of tritium of course was zero because there was no activity in the soil. The model is insensitive to crop density and evapotranspiration, as least when no transfer of activity occurs.

The transfer between TFWT and OBT in the plant occurs during what is called photosynthesis but is really just exchange based on specific activity coupled with a rate based on residence time:

$$\text{Bq TFWT/kg fw x (g OBH/kg fw / (g } H_2O/\text{kg fw x 1/9) / residence time) = Bq OBT/kg fw}$$

Here OBH is organically bound hydrogen and the factor 1/9 is the number of grams hydrogen per gram water.

TFWT is lost from the plant to a losses compartment at a turnover rate of 1 per hour during the daytime and a fraction of the daytime rate (0.06) at night.

OBT is lost to the TFWT compartment via catabolism with a rate based on the inverse of the residence time (contents of the OBT compartment are divided by the residence time of OBH).

The only difference in the way STAR-H3 handles leaves compared with flowers or fruits is by the water and hydrogen contents, or by changes in turnover rates and residence times. Leaves, shells, and beans were therefore modelled separately, although turnover rates and residence times were the same for all (given the uncertainty) (see Table 1).

The parameter values used for the soybean scenario and their distributions are shown in Table 1.

Table 1. Parameter values and distributions varied in STAR-H3

Name	Units	Best estimate	Distribution	Range
For leaves & stems				
Plant_water	g H_2O/kg fw	869	Normal	± 5.2
Hydrogen_amount	g OBH/kg fw	9.1	Normal	± 0.71
For beans				
Plant_water	g H_2O/kg fw	80	Normal	± 7.4
Hydrogen_amount	g OBH/kg fw	72	Normal	± 2.2
For shells				
Plant_water	g H_2O/kg fw	90.3	Normal	± 10
Hydrogen_amount	g OBH/kg fw	58	Normal	± 5
Parameters for all				
Water_turnover_day	h^{-1}	1	Uniform	0.5 – 2.0
Water_turnover_night	frac. Day value	0.06	Triangular	0.01-0.06-0.1
OBT_Residence_time	d	39	Normal	± 9.3

Table 2 shows the water contents used for the parts of the soybean plant to convert predicted concentrations in Bq/kg fresh weight to Bq/L (and to calculate the concentrations in Bq/kg fw in STAR H-3 from air moisture concentrations).

Table 2. Parameter values to convert Bq/kg fresh weight to Bq/L

	leaf	shell	bean	pod
Fresh matter fraction	0.869	0.0903	0.08	0.085
Dry matter fraction	0.131	0.9097	0.92	0.915
Water equivalent	0.6	0.59	0.7	0.65

3. PREPARATION OF INPUT

Absolute humidity was calculated for each 5-minute period using the 5-minute observed relative humidity and temperature for each experiment. Then, using the 5-minute calculated absolute humidity and the observed 5-minute air moisture concentrations (Bq/mL), Bq/m^3 for each 5-minute period was calculated. The air concentrations for the 13 time periods of each experiment were averaged for the hourly input to STAR-H3, and the mean absolute humidity was obtained from averaging the calculated 5-minute absolute humidity for the 13 time periods (see Table 3). The uncertainty on the mean absolute humidity was assumed 10% of the value, disregarding the rapid rise and fall of absolute humidity at the start and finish of the experiment. Table 3 also shows the number of hours of each run and the number of runs for the stochastic output. Note that the air concentration for STAR-H3 is deterministic.

Table 3. Input to STAR-H3

	Air Bq/m3	AH (g m^{-3})	Hours	# Runs
SB1	3.30 E+06	41.4 ± 4.14	2280	1000
SB2	4.46 E+06	29.6 ± 2.96	2016	1000
SB3	4.82 E+06	41.4 ± 4.14	1608	1000
SB4	2.69 E+06	50.5 ± 5.05	1368	1000
SB5	3.68 E+06	37.8 ± 3.78	1008	1000
SB6	3.80 E+06	27.6 ± 2.76	432	1000

Day length was adjusted based on approximate hours of daylight at Seoul for the day of each experiment. Of course, actual day length got shorter before harvest, which was not taken into account.

SB1: sunset at 20:00; sunrise at 5:15; midpoint of experiment: 10:00
SB2: sunset at 20:00; sunrise at 5:15; midpoint of experiment: 10:00
SB3: sunset at 19:45; sunrise at 5:30; midpoint of experiment: 10:15

SB4: sunset at 19:30; sunrise at 5:45; midpoint of experiment: 10:00
SB5: sunset at 19:15; sunrise at 6:00; midpoint of experiment: 9:30
SB6: sunset at 18:45; sunrise at 6:15; midpoint of experiment: 10:00

It was assumed (incorrectly, as it turned out), that unless the shell or bean were growing at the time of exposure, no significant amount of tritium would be transferred. STAR was therefore used to calculate hourly concentrations (with modifications, see below) for only those endpoints shown by the X's in Table 4.

Table 4. Parts of the plants that were growing when exposed to tritium.

	Leaves and stems	Shells	Beans
SB1	X		
SB2	X		
SB3	X	X	
SB4	X	X	X
SB5	X	X	X
SB6	X	X	X

4. MANIPULATION/ADJUSTMENT OF STAR RESULTS IN EXCEL

As mentioned, STAR-H3 basically assumes rapid equilibrium of the final product (mature leaf, mature bean) at the time of exposure. However, in these experiments, growth was occurring. Therefore, a loss was applied to all the hourly output data from STAR-H3 based on estimated growth rates (Table 5) – as the plant doubled in size, the concentrations of tritium were halved. These growth rates were estimated from the KAERI data.

Table 5. Growth rate (doubling) in days of parts of soybean plant

	Fastest growth	est estimate	owest growth
Leaves and stems	50	55	60
Pods	5 until Sept 2, then ∞	30	45
hells (after July 12)	40	45	50
eans (after July 24)	0 to Aug 24, then ∞	30	40

For those parts of the plant not growing when exposed to tritium (not marked with an X in Table 4), it was assumed that the starting concentration in the pod or bean was the same in Bq/L as the concentration in the leaves on the day the pod or bean started to grow (shells were assumed to start growing July 12; beans, July 24). The STAR loss rate from shell or bean was

then applied to the new concentration derived from the HTO concentration in leaves. This approach did not account for differences in concentrations between day and night in STAR (which are quite large but never entered into this scenario because all concentrations were measured in daytime[1]). Obviously, there was just a tiny amount of TFWT in the leaves (by these calculations) when shells and beans started to grow.

6. ESTIMATION OF UNCERTAINTY USING CRYSTAL BALL® RISK ASSESSMENT SOFTWARE

Some sources of uncertainty are not accounted for by STAR-H3. To account for one of these additional sources of uncertainty (in the source term) the air concentrations ± one standard deviation and the absolute humidity ± one standard deviation for each scenario (Table 6) were multiplied together in the Crystal Ball® Risk Assessment Software to (re)calculate the air concentrations and calculate the percent associated uncertainty. All distributions were considered normal.

Table 6. Input to Crystal Ball to determine the uncertainty on the initial air moisture concentrations

	Mean Air (Bq/L)	Standard deviation	Mean absolute humidity (kg/m3)	Standard deviation
SB1	$8.35\ 10^{7}$	$1.31\ 10^{7}$	0.0414	0.00732
SB2	$1.59\ 10^{8}$	2.3710^{7}	0.0296	0.00336
SB3	$1.23\ 10^{8}$	$2.95\ 10^{7}$	0.0414	0.00370
SB4	$5.61\ 10^{7}$	$1.54\ 10^{7}$	0.0510	0.00874
SB5	$9.91\ 10^{7}$	$2.55\ 10^{7}$	0.0378	0.00805
SB6	$1.48\ 10^{8}$	$5.71\ 10^{7}$	0.0284	0.00712

Air concentrations in Bq/m3 and percent uncertainty (1 standard deviation) predicted by Crystal Ball are shown in Table 7. The median air concentrations predicted by Crystal Ball (Table 7) were within about 5% of the deterministic air concentrations used as input to STAR.

1 The STAR-H3 model output exhibits much higher concentrations at night than during the day. For example, for experiment SB1, the highest concentration from STAR output in any 24 hour period occurs at 5:00, and the lowest occurs at 20:00; the ratio of the highest divided by lowest concentration for each 14 hour period is 8.9! Note that these extreme values occur when the loss rate of the plant changes from day to night and vice versa. The concentration taken as the prediction for this scenario was at 10:00. This value is just 3% higher than the lowest concentration and only 11% of the highest concentration. This behavior is inexplicable, because, although the loss rate from the plant slows at night, the water content must remain about the same so there can be nothing driving an increase in concentration after an acute exposure.

Table 7. Air concentrations and standard deviations as calculated by Crystal Ball

	Median (Bq/m^3)	Standard deviation (Bq/m^3)	Percent uncertainty
SB1	$3.46\ 10^6$	$6.11\ 10^5$	17.7
SB2	$4.65\ 10^6$	$8.79\ 10^5$	18.9
SB3	$5.04\ 10^6$	$1.32\ 10^6$	26.2
SB4	$2.81\ 10^6$	$9.36\ 10^5$	33.4
SB5	$3.62\ 10^6$	$1.26\ 10^6$	34.9
SB6	$4.07\ 10^6$	$1.95\ 10^6$	47.9

Another source of uncertainty not taken into account by STAR (which assumes the tritium-to-hydrogen (T/H) ratio is maintained throughout the environment) is the empirical reduction in the T/H ratio between air moisture, leaves and fruits and in the T/H ratio between TFWT and OBT. This reduction of T/H ratio was described using triangular distributions (Table 8). The uncertainty on the distribution for pods is quite large because, when the soil is not contaminated, the T/H ratio is often observed to be low in equilibrium conditions.

Table 8. Triangular distribution of reduction factors between air moisture and TFWT and TFWT and OBT

	Lower limit	Midpoint	Upper limit
HTO in leaves	0.5	0.75	1.0
HTO in pods	0.2	0.5	0.9
OBT in leaves	0.4	0.6	0.8
OBT in pods	0.1	0.3	0.5

There's additional uncertainty on when shells and beans start to grow and whether or not they can be exposed directly to the HTO or what the concentration in the plant is at the start of growth. This affects experiments SB1 and SB2. The uncertainty is expressed as the fraction of time on either side of the assumed initiation of growth dates (July 12 for shells and July 24 for beans).

SB1 shell: uniform 0.937 – 1.13

SB1 bean: uniform 0. 82 – 1.2

SB2 shell: uniform 2.2 10^{-3} – 1.7 10^{4} (note extreme uncertainty)

SB2 bean: uniform 0.82 – 1.2

To calculate the effect of these additional sources of uncertainty on the predicted concentrations in parts of the plant, the mean of each STAR (or Excel-massaged STAR) air concentration at time x, with the 2.5 and 97.5% values of the distribution (assumed lognormal) obtained from STAR, was multiplied by 1 ± percent standard deviation for the uncertainty on the air concentration during the experiment (Table 7) times the triangular reduction factor distributions (Table 8) times the ranges on uncertainty on times the growth started. Each distribution was sampled 5000 times. The outcome for each experiment was a new mean with new 2.5% and 97.5% confidence limits.

In nearly all cases, the uncertainty on the results increased after running Crystal Ball.

7. DISCUSSION AND EXPLANATION OF RESULTS

The average estimated air moisture concentrations calculated were all within 5% of the observed, so the starting air concentration for each experiment was not a cause of any differences between predictions and observations. Results are presented as predicted-to-observed (average) ratios in Tables 9a and 9b.

Table 9a. P/O ratios for HTO in leaves and pods.

SB1			SB4		
Time in hours	Leaf/stem	Pod	Time in hours	Leaf/stem	Pod
1	17		1	1.8	0.63
24	0.24		24	0.041	0.00039
120	0.010		120	0.0029	0.0047
336	0.025		336	0.0049	0.12
936	0.025	0.132	768	0.0065	0.083
1608	0.014	0.149	1368	0.0043	0.082
2280	0.0042	0.061			

For leaves and stems, the observed concentrations in SB4 are higher both absolutely (more than a factor of 2, and sometimes much more) and relatively (because the air moisture concentration for SB4 is about two-thirds that of SB1) than those of SB1. Given that the model behaves the same way for SB1 and SB4, the differences in the P/O ratios are due to the differing dynamics of the observations. These variations aside, the dynamics are very different in the model compared with the observations, with over-predictions in the first hour followed by more-or-less increasing under-predictions with time.

Predicted HTO concentrations, particularly towards harvest, are lower than those calculated by STAR alone due to the introduction of growth. For SB1, the HTO concentration in leaves and stems at 2280 hours was one-third that of STAR; for SB4, it was about half that of STAR. Obviously, by introducing growth, the differences between predictions and observations became greater than they would have been had STAR results been used. Furthermore, as pointed out in footnote 3, the daily dynamics of STAR do not make sense. Any night time concentration would be significantly higher than the one reported for 10 am and, if chosen, would further decrease the large discrepancy between predictions and observations. Of course, there is no reason to support this action, but then, there seems to be no reason for the large hourly fluctuation in concentrations.

Predicted concentrations of OBT at harvest get closer and closer to the observations as the time between experiment and harvest becomes smaller (Table 9b) (concentrations are highest at 432 hours and lowest at 2016 hours). This implies that the residence time in STAR is shorter than in the experiment and that the turnover time for shells and beans is faster than in the experiment; the result is that predictions and observations diverge over time. Furthermore, my model does not account for the fact that, in nature, tritium (or any nuclide) is taken up at a higher rate at certain stages of growth, which is seen in the experimental data in which OBT concentrations in experiments SB4 and SB5 are higher than those in experiment SB6. The P/O ratio for SB2 shells is relatively very high. This is because of the enormous uncertainty applied for when the shells started to grow compared with time of exposure.

Table 9b. P/O ratios for OBT in leaves, shells and beans

	Leaves	Shells	Beans
SB1 (2280 h)	0.095	0.00029	0.00011
SB2 (2016 h)	0.059	0.18	0.000052
SB3 (1608 h)	0.12	0.012	0.012
SB4 (1368 h)	0.26	0.011	0.0089
SB5 (1008 h)	0.46	0.039	0.034
SB6 (432 h)	0.52	0.42	0.41

Observations fell within uncertainty bounds in just 2/40 cases. In another 5 cases, one of the observations (e.g., for stems or leaves) fell within uncertainty bounds. In 33/40 cases, the observations were outside the uncertainty bounds. The average magnitude of the uncertainty (disregarding huge uncertainties generated at 24 hours by STAR was a factor of 41 (value of 97.5% confidence limit (CL) divided by value of the 2.5% CL), with a range of 3.3 (HTO in leaves and stems for SB1) to 486 for OBT in beans for SB1. For those cases where the observation fell within the uncertainty bounds, the magnitude of the uncertainties was less than a factor of 13, so when the model was right, it was confidently right (although probably mostly by chance), because the fewer hours between the experiment and harvest, the better the model did.

8. CALIBRATION OF STAR

Based on the results it was considered possible that STAR could be calibrated to resemble the observations (ignoring uncertainty). Only three parameters can be changed – the turnover time of tritium in the plant (day and night) and the residence time of tritium in the plant. Without attempting to change the night/day default water turnover rate in STAR, the other two parameters for HTO and OBT in SB4 leaves and pods were varied. The best results were from:

- For leaves, changing the turnover rate from 1/h to 0.25 per hour and leaving the residence time at 39 days.
- For pods, changing the turnover rate from 1/h to 0.1 per hour and leaving the residence time at 39 days.

This resulted in P/O ratios (Table 10a) that may be compared with those from Table 9a.

Table 10a. P/O ratios for calibrated STAR for HTO (SB4)

Hours	P/O leaves and stems	P/O pods
1	1.7	0.66
24	3.1	0.48
120	0.02	0.082
336	0.042	1.3
768	0.08	1.9
1368	0.09	4.6

Overall this is much better than the original submission, but the dynamic, even without growth, still is not achieved. Note that these results do not account for any growth or reduction in the T/H ratio between air moisture, TFWT, and OBT.

The calibration had to be done with Experiment SB4 because only it had HTO in pod data for the full time period. The OBT in pods was part of the calibration, although results were not in close agreement for SB4. Similarly (Table 10b) new OBT concentrations were calculated for beans using the changed turnover rates that can be compared with Table 9b.

Table 10b. P/O ratios for calibrated STAR for OBT

	P/O beans
SB1	28
SB2	9.4
SB3	0.65
SB4	0.17
SB5	0.42
SB6	2.7

Over-predictions for SB1 and SB2 are because STAR had the beans growing at time of exposure, which was not the case. P/O ratios increase from SB4 to SB6 because STAR assumes equal uptake of tritium throughout all stages of development.

By calibrating STAR and ignoring any growth and any reduction in the T/H ratio, the predictions are greatly improved but the dynamic is still unattained. Calibrating STAR also may have reduced the turnover rate below a reasonable value.

Although STAR results could not duplicate the dynamics of the experiment, how well the integrals over time could be predicted was investigated. The estimated observed integrals (hourly sums) for HTO are compared in Table 11 in stems and leaves with the integral of mean predictions, both as submitted and as calibrated.

Table 11. Hourly sums (Bq/mL) over length of experiments for HTO concentrations

Experiment	Observed integral	Predicted integral	P/O Ratio	STAR calibrated integral	STAR calibrated P/O ratio
SB1 leaves	27300	116000	4.3		
SB1 stems	18000	116000	6.4		
SB4 leaves	82400	55600	0.67	71400	0.87
SB4 stems	13300	55600	4.1	71400	5.4
SB4 shells	276000	55800	0.20	89400	0.33
SB4 beans	274000	55800	0.20	89400	0.33

Except for the high integral for SB4 leaves, the results of leaves and stems are very similar. The model and the calibrated STAR (no growth) model over-predict concentrations in leaves and stems, so, if ingestion is instantaneous in the model, doses will not be under-predicted (if the diet could be composed of soybean leaves!). Note that, although the calibration makes enormous improvement in the P/O ratios at harvest (compare Tables 9a and 10a), the change in the integrals from original to calibrated predictions is due primarily to the reduction in T/H ratios (original integral is 75% that calibrated for leaves and 50% that calibrated for shells and beans). This is because the extraordinarily high initial values for the first few hours dominate the entire integral; for SB1 at 2280 hours, the concentration in the soybean leaves that "grew" is 30% that of STAR's (that didn't grow).

9. CONCLUSIONS

There are just three parameters in STAR that can be changed in any attempt to calibrate the model output to the observations: the daytime turnover rate, the fraction of the daytime turnover that occurs at night, and the residence time of OBT. For HTO, the turnover rates dominate the dynamics of the predictions, but the residence time does have a small effect on the dynamics. Calibration of the HTO dynamics was attempted in STAR two ways, one described above and one in which the nighttime turnover fraction was set to 1 to simplify the output. Changing the nighttime turnover fraction to 1 did not noticeably affect the dynamic

response at the sampling time of 10 am, but of course it did eliminate that odd fluctuation in concentration mentioned in the footnote. To calibrate the model to the time-dependent HTO results alone was attempted, because to calibrate single points for OBT at harvest would be, of course, meaningless. Thus, as mentioned above, the dynamics of the STAR predictions are much improved through calibrating the model to a lower turnover time but maintaining the 39-day residence time. The calibration is fairly meaningless, because it does not include lower concentrations due to plant growth or the expected reduction in T/H ratio between compartments.

Apparently, STAR is too simple to account for the changing dynamics of the HTO concentrations. Furthermore, STAR has no way to predict concentrations in pods that are exposed while still flowers; STAR has no mechanism of uptake by the leaf or flower and consequent transport into the fruit. The OBT concentrations cannot be predicted because STAR does not recognize that uptake may be preferential during certain stages of plant growth.

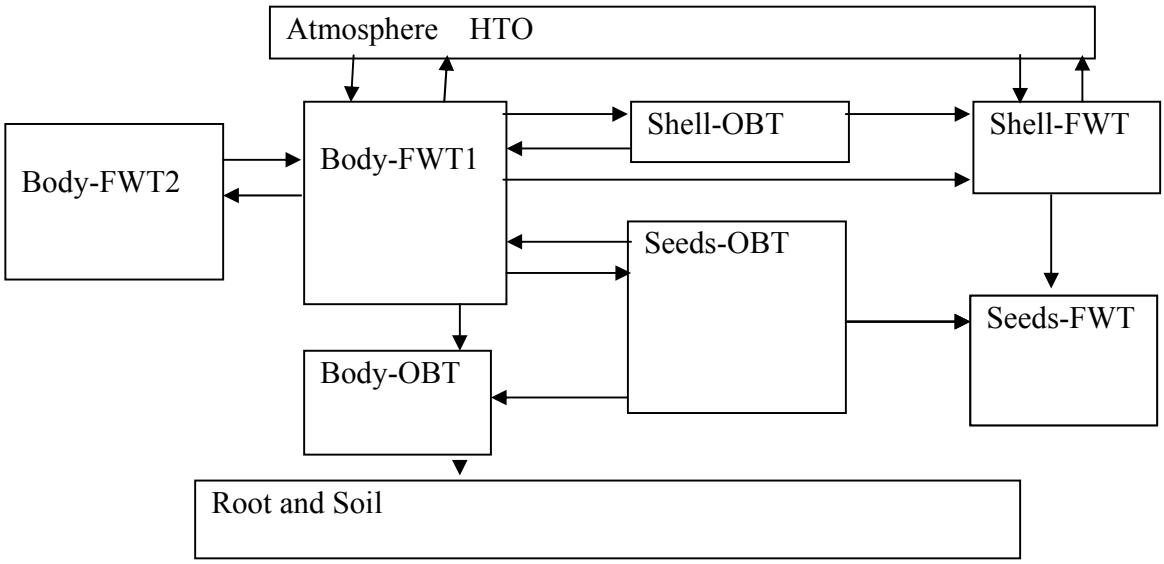
Kyoto University Model Description and Results of Calculation

1. TriSoy MODEL

TriSoy (**Tri**tium Behavior in **Soy**bean) is a simple analytical code written in Visual Basic.NET. The calculation is performed through graphical use interface and the result is implicated in an Excel spread sheet. The purpose of the model is to calculate the specific activities of FWT and OBT contained in tissues of a soybean plant that was exposed to atmospheric tritium vapor at various growth stages. The model is applicable to other crop plants by changing basic parameters used in the model.

2. STRUCTURE OF THE MODEL

The scheme of TriSoy is shown schematically below.



The main features of this model are as follows.
1) The source of tritium is solely the atmospheric HTO vapor.
2) The atmospheric tritium is taken up by the body, i.e. leaves and stem.
3) The body FWT could be transferred to the shell FWT compartment.
4) Carbohydrates are photosynthesized in the leaves and instantaneously translocated to seeds and shell.
5) A portion of OBT in the plant is converted to FWT by respiration.

3. DETERMINATION OF THE EXCHANGE VELOCITY CONSTANT

In this section, the following abbreviations are used.

Λ: Tritium exchange velocity constant
C_a : Tritium specific concentration in the atmosphere (Bq/Kg)
C_S : Tritium specific concentration in the stomata vapor (Bq/Kg)
C_L : Tritium specific concentration in the leaf vapor (Bq/Kg)
ρ_s : Saturated vapor density of the atmosphere (Kg/m^3)
r : Resistance of water vapor transfer at the leaf surface (/m)
F : Tritium flux on the leaf surface ($Bq/m^2.s$)
μ : Water content /leaf or canopy area (Kg/m2)
α : Hydrogen isotope separation factor for tritium : 1.104 at 25□
h_r : Relative humidity of the atmosphere

3.1 Resistance of water vapour transfer at the leaf surface

The process of tritium intake by the plant basically follows the concept of Belot's model (Belot 1979). The exchange velocity constant λ is given by

$$\lambda = \frac{\rho_s}{\alpha \mu r} \quad (1)$$

The value of r was determined by using the relationship between biomass production rate and transpiration rate as follows.

The dry mass production velocity ($kg/m^2.d$) is given by

$$\frac{dW}{dt} = K_w \frac{E}{\delta} \quad (2)$$

where W = amount of dry mass (kg/m^2)
K_w= conversion factor = 0.005 kPa for soybean (Cropsyst Manual), and
δ= deficit of water vapor pressure (kPa)

The transpiration rate is related to the deficit water vapor concentration (DVC) in the air as

$$E = \frac{DVC}{r}$$

or

$$E = \frac{\rho_s - \rho}{r} \quad (3)$$

where
ρ_s = saturated water vapor concentration in the air (kg/m^3)

ρ = water vapor concentration in the air (kg/m^3), and
r = the resistance of exchange of HTO and H_2O between the air and the soybean leaf. (d/m)

In principle, by using the above two equations, the value of r can be estimated.

For a representative period of the soybean growth, in the period from July 1 to August 24, the averaged dry mass production rate was 0.0224 $g/(m^2.d)$ and the averaged water vapor deficit was 0.464 kPa or 2.96×10^{-3} kg/m^3. The transpiration rate in this period was then 2.07 $kg/(m^2.d)$.

The value of the resistance r depends on the physiological condition and the meteorological conditions for the plant. As a representative weather condition, the daytime length was assumed to be 13 hours and the night time 11 hours. Then the day-averaged transpiration rate E_{aver} is given by

$$\mathbf{E_{aver} = \frac{13}{24} \bullet \frac{DVC_{day}}{r_{day}} + \frac{11}{24} \bullet \frac{DVC_{night}}{r_{night}}}$$

Further, it was assumed that in the night time the air vapor is close to saturation and the stomata are closed. Then the second term of the above equation can be neglected.

Thus,

$$\mathbf{E_{aver} = \frac{13}{24} \bullet \frac{DVC_{day}}{r_{day}}} \quad \text{or} \quad \mathbf{r_{day} = \frac{13}{24} \bullet \frac{DVC_{day}}{E_{aver}}} \qquad (4)$$

Again here, using the values of DVC_{day} and E_{aver} for the growth period of soybean, the value of r in daytime is approximated by

$$\mathbf{r_{day} = \frac{13}{24} \bullet \frac{2.96 \times 10^{-3} (kg/m^3)}{2.07 (kg/m^2.d)} = 7.75 \times 10^{-4} (d/m) \text{ or } 66.9 (s/m)}$$

The above argument is based on the plant canopy area.

To consider the resistance in individual leaves, a correction by the leaf area is necessary. Under the assumption that the transpiration velocity is in proportion to the total leaf area, the resistance of individual leaves r_i is given by

$$\mathbf{r_l = LAI \bullet r_{day}} \qquad (5)$$

where LAI= the leaf area index (LAI)

According to Tohachi, for a typical Japanese soybean species, the LAI value is 4.5.

If this value is used, the value of the resistance becomes 3.0 s/cm, being close to the value determined by Garland and Cox for dwarf French beans.

3.2 Tritium exchange rate velocity constant

Under some situations, the release of tritium to the atmosphere from the plant leaves should be taken into consideration as well. In the present scenario, the plant body FWT quickly equilibrates with the atmospheric HTO vapor. Therefore, the HTO level of the plant water can be at the same level in order throughout the exposure time. In such a case, release of the existing HTO from the plant leaves could not be negligible. Then, the tritium concentration in the leaves at time t or $C_L(t)$ is given by

$$C_L(t) = C_L(0)\exp(-\frac{\rho_s t}{\alpha\mu r}) + (\alpha h_r C_a)\left(1 - \exp(-\frac{\rho_s t}{\alpha\mu r})\right) \quad (6)$$

In an extreme case of $C_L(0)=0$ the above equation is reduced to

$$C_L(t) = (\alpha h_r C_a)\left(1 - \exp(-\frac{\rho_s t}{\alpha\mu r})\right) \quad (7)$$

The exchange velocity constant λ is given by

$$\lambda = \frac{\rho_s}{\alpha\mu r}$$

Substituting the values of α and r to the above relationship,

$$\lambda = 1.29\times 10^3 \bullet \frac{\rho_s}{\mu}(/d) \quad (8)$$

or

$$\lambda = \frac{63.8\rho_s}{\mu}(/h) \quad (9)$$

Some predicted values of λ for the soybean experiment are as follows. The presented values are those averaged for the exposure time

Experiment	Date of experiment	Water content of the body (g/m2)	Water vapor density in saturated air (g/m3)	λ(/h)
SB1	2-Jul	788	47.19	2.93
SB2	13-Jul	960	30.63	1.56
SB3	30-Jul	1699	44.28	1.27
SB4	9-Aug	983	80.7	4.02
SB5	24-Aug	1647	41.31	**1.23**

SB6	17-Sep	1361	28.86	**1.04**

3.3 Dependence of the exchange rate velocity on solar radiation

During the soybean exposure experiment, the solar radiation flux changed between experiments and time to time. The intensity of solar radiation may influence the photosynthesis of organic compounds in the plant. The λ values for individual time steps were determined by considering the solar radiation flux as follows.

The biomass growth rate is proportional to the effective flux of solar radiation.

$$\frac{dW}{dt} = C_s S_a \qquad (10)$$

where C_s = light to biomass conversion factor (kg/MJ)
S_a = flux of solar radiation (MJ/m^2).

Let λ_0, S_{a0} and h_{r0} to be the values of λ, S_a and h_r averaged for the whole growth period, correspondingly.

By using the relationships (1), (2) and (10), λ is related to λ_0 as

$$\lambda = \frac{\lambda_0 \delta S_a h_{r0}}{\delta_0 S_{a0} h_r} \qquad (11)$$

3.4 OBT production

Production of OBT was assumed to take place only through photosynthesis. Let the specific activities of FWT and OBT, amount of biomass, FWH concentration of the biomass (w/w) at time t to be F(t), C(t), B(t) and γ, respectively.

From the relationship $d(\gamma B(t)) \cdot F(t) = (\gamma B(t))dC(t)$ the specific activity increase of OBT is described by

$$dC(t)/dt = (F(t)/B(t))(dB(t)/dt) \qquad (12)$$

3.5 Tritium exchange rate at pods

The HTO exchange rates in the pods and the seeds were assumed to be 1/30 and 1/15 of that in the leaves respectively.

3.6 Growth rate of biomass

In the present calculation, the information on the growth rate of the biomass at any time is necessary. Therefore, the scenario data were processed by a data processing software S-PLUS to approximate the real growth curve by a logistic growth curve. For instance, the growth curve of the body dry weight $B_d(g/m^2)$ is given by

$$B_d = 735/(1+3.5\exp(-0.079\times(t-41))$$

where t = the time elapsed after HTO exposure (d)

3.7 Tritium retention after HTO exposure

According to Ichimasa et al. (2003), after exposed to heavy water vapor the plant body heavy water taken up by exchange process is released slowly with a rate constant that is higher than that for the initial take-up process. This means that there are at least two free water compartments in the plant body. In the present model, two FWT compartments were included. Referring to the result of Cline using French dwarf bean (Cline 1953), the pool size of the second compartment was assumed to be 1.5 % of the whole free water and the retention rate constant 0.00055 d^{-1}.

Under the present scenario, the fraction of FWT converted to OBT was estimated to be less than 1%. The amount of the FWT produced by oxidation of OBT was neglected.
In night, HTO exchange velocity is considerably small compared with that in the daytime. By using the values presented for a heavy water experiment, the value of λ in night was assumed to be 1.1 (h^{-1}).

4. RESULTS AND COMPARISON WITH OBSERVED RESULTS

The main features of the model prediction are as follows.

1） The FWT level after exposure will decay to the BG level within 2 weeks.
2） The production rate of OBT is high in growing organs and tissues.
3） The translocation rate (TRL) after one-hour exposure is in the order of 10^{-3}.
4） In Experiment 6, tritium incorporation into organic material is of miner importance in OBT production since the plant is not growing supposedly.

A considerable discrepancy was seen for the FWT component of the soybean organs after exposure. The predicted FWT concentration in the soybean body at 0.2 hr after exposure was about 4 times larger than the observed value. The same tendency was seen for the results of other modelers. A possible reason of this discrepancy may be due to the tritium exchange velocity that was considerably low compared with that actually found from the laboratory experiment. Seemingly a value of λ around 0.3 (h^{-1}) is necessary to explain the observed FWT concentration. The reason of such low efficiency of HTO exchange is unclear.

In the present model, the pool size of the second FWT compartment was 1.6 % of the first compartment. But the actual contribution from the second compartment was by one order of magnitude less than this as was revealed by the Korean experiment.

Concerning OBT concentration, the model prediction for the pods from 3 to 5 agreed rather well with the experimental result. This fact validates the model for OBT production based on biomass growth kinetics. For the pods 1 and 2, the OBT translocation to the pods from other plant tissues was neglected. However, the experimental result clearly shows that such translocation of OBT should be also taken into consideration.

5. REFERENCES

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RFNC-VNIIEF Model Description

1. INTRODUCTION

The model is based upon the data given in the paper Michiko Ichimasa, Caiyun Weng, Tetsuki Ara and Yusuke Ichimasa, "Organically bound deuterium in rice and soybean after exposure to heavy water vapor as a substitute for tritiated water", Fusion Science and Technology, vol. 41 May 2002, p. 393-398.

2. MODEL DESCRIPTION

2.1 HTO uptake during exposure.

The following equitation was used for modeling of HTO accumulation during the exposure:

$$C_{HTO}(t) = C_{MAX} \cdot [1-\exp(-K \cdot t)] \qquad (1)$$

where C_{HTO} is HTO concentration in a plant sample at time t;
C_{MAX} is the steady state concentration; and,
K is the rate constant.

Table 1 shows C_{MAX} and K of our model.

Table 1. C_{MAX} and K

Plant part	C_{MAX}, relative units	K, hr^{-1}
Stem	0.534	0.069
Leaves	0.562	2.951
Shell	0.534	0.069
Seeds	0.273	0.23

2.2 HTO loss after exposure.

HTO loss equation is the following:

$$C_{HTO}(t) = C_0 \cdot \exp(-K \cdot t) \qquad (2)$$

where C_{HTO} is HTO concentration in a plant sample at time t;
C_0 is HTO concentration in air moisture; and,
K is the exchange rate.

Table 2 shows K of our model.

Table 2. K

Plant part	K, hr^{-1}
stem	0.347
leaves	1.058
shell	0.139
seeds	0.139

3. RESULTS

3.1 OBT at harvest.

OBT concentration was calculated as part of HTO concentration in air moisture. At that the plant growth phase was taken into account.

Table 3 shows the ratio of OBT concentration to HTO concentration.

Table 3. Relative OBT concentration to HTO concentration in air moisture

Experiment	Plant growth phase	Ratio
SB1	The beginning of the growth.	$5 \cdot 10^{-5}$
SB2	growth phase	$1 \cdot 10^{-4}$
SB3	growth phase	$1 \cdot 10^{-4}$
SB4	growth phase	$1 \cdot 10^{-4}$
SB5	growth phase is finished	0.0
SB6	growth phase is finished	0.0

EDF Model Description and Discussion of Results

1. MODEL DESCRIPTION

The EDF model used to calculate tritium concentrations in crop was developed for continuous release. It required to be adapted to cover the soybean scenario. The main assumptions made are described here:

- Fluxes of HTO from air to plant leaves were calculated according to Belot's equation (Belot, 1979). A five-minute time step was used; concentrations were assumed to be constant over the time step and equal to the concentration measured at the end of the time step. The value assigned to the exchange rate during the day was 1mm/s, except in SB4 where the value was twice lower to take into account the effect of the high temperature and the low relative humidity on stomatal closure. At night the exchange rate was assumed to be 50 times lower than during the day.
- Background HTO concentration in atmospheric water vapour was calculated from the average monthly tritium concentration in air and meteorological data. The average value was $2.4\ 10^{-3}$ Bq/mL.
- OBT formation during each time step is proportional to the growth rate and to the concentration of tritium in the tissue free-water. A discrimination factor of 0.6 is used (ratio between T/H in OBT and T/H in HTO). OBT is calculated with a daily time-step. Linear growth rates on a dry weight basis were derived from the soybean experimental data: 7.5 $g.m^{-2}.day^{-1}$ for shoot and 7.7 $g.m^{-2}.day^{-1}$ for pods. The same growth rates were applied in all experiments.
- OBT conversion to HTO is not considered. Thus, decrease in OBT concentration is only due to dilution by uncontaminated dry matter formed after exposure.

2. DISCUSSION

From the results of this experiment, it seems that OBT conversion to HTO should be included in the model. What was considered at first to be a conservative assumption is shown to under estimate the HTO concentration in the plant free water in the post exposure phase and consequently the OBT concentration in the pods when fruit formation starts after exposure.