Overview on tritium transfer from air to plants and conversion to OBT with focus on night cases

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I. INTRODUCTION

Tritium, as an isotope of hydrogen, enters into many organic forms, but the interest for nuclear safety resides in its bio-available forms [1]. The Organically Bound Tritium (OBT) is defined as: "OBT is the activity in the combustion water of dry bio-matter that has been washed repeatedly with tritium free water. It represents carbon-bound tritium and buried tritium that was originally formed in living systems through natural environmental or biological processes from HTO (or HT via HTO)" [2] and is primarily formed through photosynthesis in plants, in the presence of tritiated water in leaves. During the night, the specific metabolic processes are involved. OBT concentration in crops is the essential information necessary for assessing ingestion doses. Its "non-exchangeable' form is of primary interest, because its dose coefficients are about 3 times higher than for tritiated water.

OBT generation in the darkness has already been observed by Moses and Calvin [3] who exposed chlorella algae to HTO in their nutrient solution under conditions of light and darkness for 3 min. The tritium incorporation into non-exchangeable positions of the organic matter in the dark was one-third of that in the light. Thompson and Nelson [4] exposed primary leaves of soybeans to HTO in the atmospheric humidity under conditions of light and darkness for 1 or 30 min. Related to the same exposure time, the assimilation of tritium in the dark was only 10 % of that in the light.

While formed in leaves, OBT is translocated in the edible plant parts, most of which are reproductive organs, and depends on the growth stage of the plant at the time of exposure. OBT concentration in the edible plant part is highest in the generative period when the fruits grow [5,6].

For ingestion dose, the contribution of OBT was first introduced in 1994 [7], but officially only recently was considered in the Canadian Standard for routine releases [8].

For routine releases, the contribution of OBT for the ingestion dose is less than 30 % and highly depends on local consumption. For accidental releases, there is still a debate and in some situations, it can be as high as 80 % of the tritium dose [9]. Until now, there was not possible to harmonize different views and to agree on the best model for risk assessment due to accidental tritium release. The requirements for a robust, transparent and relatively simple model, but still moderately conservative, were formulated, but the development of such a model is difficult, because tritium transfer to crops is subject of changing environmental (meteorology) condition and depends on many plant physiological processes with site specific parameters (adaptation). Models must be based on recognized physical basis and must be subjected to quality assurance procedures including uncertainty and sensitivity studies, but also tested with experimental data [10]. Few blind tests of models with the experimental data have been done in the past [11, 12] and this limits our performance in demonstrating the model reliability.

Concerning the impact of night production of OBT, the debate has a long story, because the experimental results show that the formation of OBT in night condition is less than in the day conditions, in the majority of situations, at similar air contamination. For the same source term, the air concentration of tritiated water concentration in night is much higher than in day conditions (up to 40 times). Uptake of HTO by leaves in night was higher than expected and can be only a third from the day uptake. Conversions of OBT in night have been observed to be higher than in day in few occasions (see later).

A recent review of tritium in plants [13] have preliminary covered the impact of OBT for ingestion dose, while reviews of French and Canadian nuclear safety authorities [14,15] concentrate more on routine releases. A detailed review on experimental findings for short term exposures is missing and it is the aim of the present report in order to do a better assessment of the potential impact of OBT under accidental events. Published, but also results not accessible in open literature, will be presented, including the night conditions. A brief revision on processes involved in the production and transport of assimilate in crops will be included but modeling approaches will be a subject of further report.

The experimental data presented is organized according with the country of origin and the key aspects are reviewed. The aim of this contribution is to point out the actual difficulties and the need of further collaboration at international scale.

II. GERMANY: WHEAT, BEAN, POTATO

In the first systematic experimental approach [16], a dedicated experimental climatic chamber was used in the laboratory and for the night experiments, and a diminished leaf uptake rate was observed, because of a significant closure of the stomata which was obviously not complete. The concentration ratios in night conditions are reduced to 23 % in leaves, to 25 % in stems and 59 % in ears, compared to those observed in high light conditions. There was no significant difference in the HTO uptake between spring wheat and winter wheat leaves. In leaves, the initial relative OBT concentrations were typically ½ in night conditions in comparison with high light conditions. It has been clearly demonstrated that there is a small, but not insignificant OBT incorporation in night conditions in leaves, stems and ears, indicating that tritium can be incorporated into organic matter not only by photosynthesis, but also by metabolic pathways independent of light, for instance by reactions of the tricarbonic acid cycle or other metabolic conversions. In an extended night experiment, the OBT concentrations in the ears increased by a factor of 3 during the extended dark period. This indicates high rates of metabolic turnover in the ears, which does not result in *de-novo* synthesis of organic material.

The total OBT increased until day 1 after exposure, because the HTO concentrations in the exposure box decreased slowly and, correspondingly, the TWT concentrations in the plants. Already after day 1, the OBT had been transported into the grains. While the percentages of OBT in leaves, stems and husks decrease with time, the grain OBT increases considerably until harvest time. It is suggested that small actual loss of OBT occurs from grain once translocation has taken place.

To quantify the translocation of OBT to grain, a so-called **translocation index (TLI) has been defined. This is the percentage of the OBT concentration in grain at harvest (Bq/ml water of combustion) related to the TWT concentration in leaves (Bq/ml) at the end of exposure to HTO. This definition will be used in the report.**

The TLI, observed in a series of exposure experiments with potted spring wheat in different growth stages between anthesis and maturity, shows that the final OBT concentration in the grain was highly dependent on the time of exposure (Table G.I and Figure G.I). In night-time conditions (experiments Wn2b WN2a) the uptake into TWT of leaves was about four times lower than in daylight conditions. Related to the same TWT concentration, the initial OBT concentration in leaves is about half of that in daylight conditions.

The absolute value of TLI in Figure G.I is not relevant, because leaves maintain high HTO level for long time in the experiment and formation of OBT in leaves is longer than 2 hours (when leaf HTO is used for defining TLI). It is important to note that the night values are close to the day ones.

Table G.I. Experimental data (Diabate and Strack, Personal communication (excel AUSW93b from Sigfried Strack) Bq/mL)

	W6b	**W7b**	**W6a**	**W7a**	**WN2b**	**W8b**	**WN2a**	**W8a**	**W9b**	**W9a**
DAF	1	6	7	12	13	15	19	21	27	33
OBT										
ear (1 d)	831	478.6	568	787		822	427	384		
ear (harvest)	81.2	113.7	151	381	**222**	523	357	481	394	296
grain (harvest)	51	229	179	483	474	657	447	610	543	362
HTO										
leaf (2 h)	81899	89379	94254	86642	**72911**	106130	**67441**	93513	98587	96886
leaf (+1h)		64347		66938	**59431**		**71115**		91702	80361
leaf (+2h)	49727		50433	52124		45824	**56562**	50115	73764	74301
leaf (1 d)	2934	4720	3903	4677		6329	6051	6383	4675	

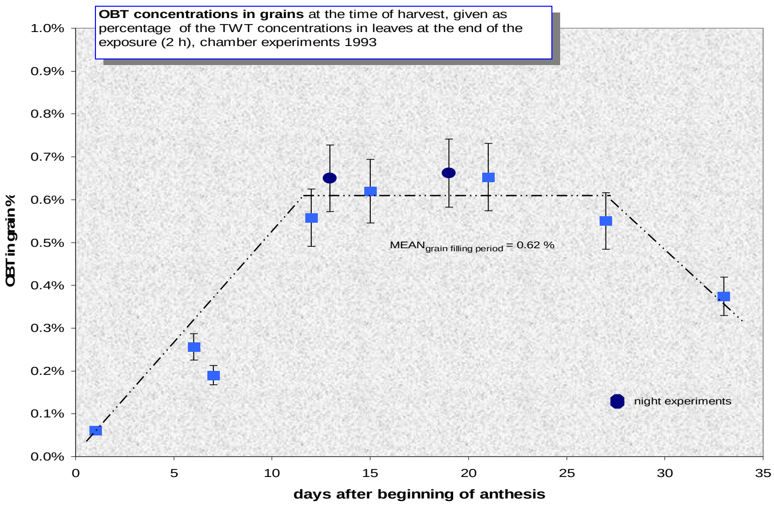


Figure G. I. Translocation index for OBT in grain(from Strack, [17])

The shape of the time dependence of the translocation index in Figure G.1 can be explained by general processes of wheat growth. If we consider the WOFOST crop growth model [18]), a generic winter wheat in Germany and the average weather in Central Germany, we obtain the results given in Table G. II, where Daa is the day after anthesis (flowering), growth of grain (dry matter – dm) is in kg/ha, and the growth rate of the grain and aboveground plant are in kg/ha d. Partition reflects the share of the new dry matter translocated to the grain (after respiration). For leaves we have LAI and the green leaves mass (dm kg/m^2). M/G is the ratio between maintenance respirations and gross assimilate production. At the beginning of the grain filling, the partition to grain is small and the growth dilution effect is high (see ear at day 1 and harvest). This explains the low TLI. At the end of the grain filing period, much of the OBT formed in the leaves is used for maintenance respiration and few remains to be translocated to grain.

Table G.II. WOFOST, winter wheat, Germany (IFIN results)

Daa	**grain (kg/ha)**	**grain rate (kg/ha d)**	**plant rate (kg/ha d)**	**partition**	**LAI (m^2/m^2)**	**WLV (kg/m^2)**	**M/G**
-2	28	28	254.2	0.1	6.91	0.326	0.28
0	309	178	261.2	0.7	6.79	0.3203	0.29
2	820	255	258	1.0	6.68	0.3152	0.29
6	1828	250	251.4	1.0	6.49	0.3063	0.31
11	3061	244	242.5	1.0	6.3	0.2971	0.33
19	4933	222	216.3	1.0	6.06	0.2858	0.36
21	5360	211	205.8	1.0	6.01	0.2836	0.37
26	6335	183	177.5	1.0	5.37	0.2534	0.41
33	7407	124	113.9	1.1	3.09	0.1456	0.45
34	7521	114	103.4	1.1	2.86	0.135	0.47
37	7799	82	70.2	1.2	2.25	0.1059	0.52
44	8012	0	0		1	0.047	0.92
56	8012	0	0		0.06	0.003	1.00

A series of experiments were done in the field, in order to observe a daily variation of the OBT accumulation in grain at harvest [17, 19]. A plastic box was mounted in field and tritium was released in the box for one hour (with ventilation). Subsequently, the box was open and the plants were left to grow naturally. The conditions in the box (relative humidity, temperature) have been recorded, as well as the photosintheticaly active radiation above the box (PPFD). The experimental data for the duration of HTO contamination in the box atmosphere are given in Table G.III, where there are reported the start hour, average temperature and relative humidity, PAR outside the box and day after flowering. Note that experiments in 1996 (bolded in Table G.III) have a better quality, because the level of CO_2 in the box was maintained at natural values.

Table G. III. The experimental data for the duration of HTO contamination in the box atmosphere [17]

Experiments	f3	**f14**	f7	f2	f4	**f10**	**f15**	f1	**f9**	**f13**	f5	**f11**	f6	**f12**
Start (h)	7	7	8	9	10	11	11	14	15	15	20	20	23	23
T (°C)	18	11	26	28	29	26	32	33	36	29	24	15	17	12
RH (%)	76	93	76	76	63	75	63	70	70	72	84	89	89	93
PPFD (µmol/m^2s)	160	179	370	644	1230	1160	1830	1180	1375	1170	54	86	0	0
DAA	18	22	24	17	18	14	28	15	12	21	22	20	22	20

The initial (1 h) uptake of HTO in the leaves relative to the average HTO in air moisture in the box is given in figure G.II

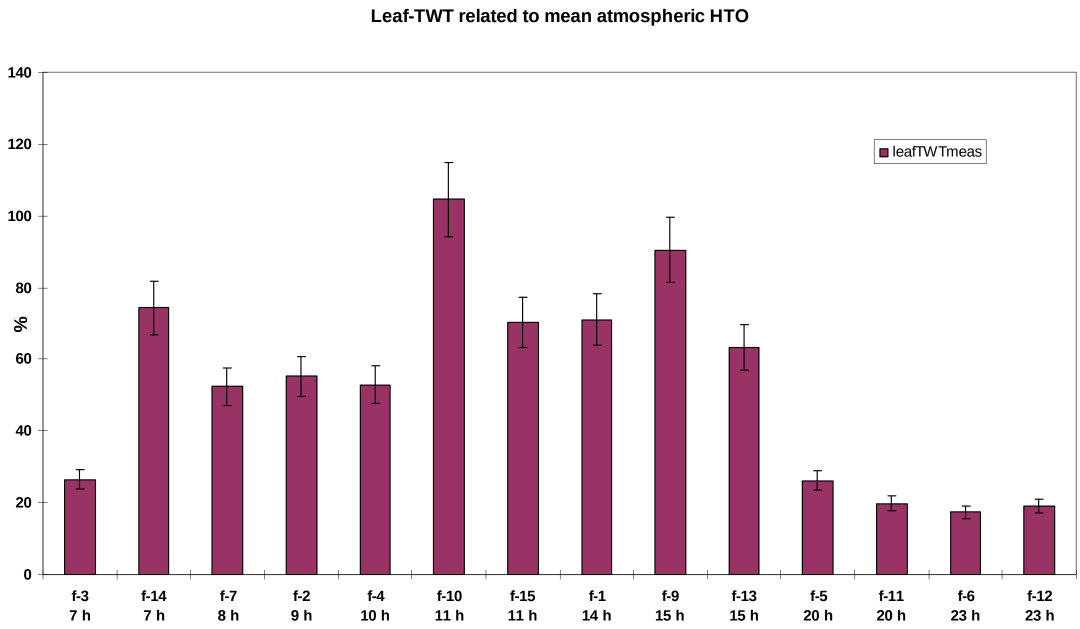


Figure G.II. Leave HTO relative concentration (relative to the average atmospheric HTO at exposure)

The mean atmospheric HTO in the box, during the 1 hour exposure, was difficult to be maintained at constant level and the mean value (as considered in Figure G.II) is not the same with values in the last quarter of hour (see Figure G.III for the dynamics of HTO in air moisture (x axis in minutes, y axis in kBq/ml)]. Considering the details on air concentration, light and temperature, the leaf HTO at the end of exposure can be successfully modeled in the frame of the actual knowledge [20, 21, 22].

The maximum relative Tissue Free Water Tritium (TFWT) concentrations were reached in the leaves in conditions of strong sunlight when the stomata were open (mean = 73 ± 19 %). The uptake was only slightly reduced in senescing leaves. In the night experiments, a diminished uptake into TFWT of leaves, stems and ears was observed because of the closure of the stomata (mean = 18 ± 1 %). The day-night difference can also be observed in stems and ears where the relative TFWT concentrations are much lower; however, the surface relative to mass is smaller than in leaves. The HTO uptake into leaves under laboratory conditions was 86 ± 2 % in high light conditions and 20 ± 7 % during the night after 2 h [19].

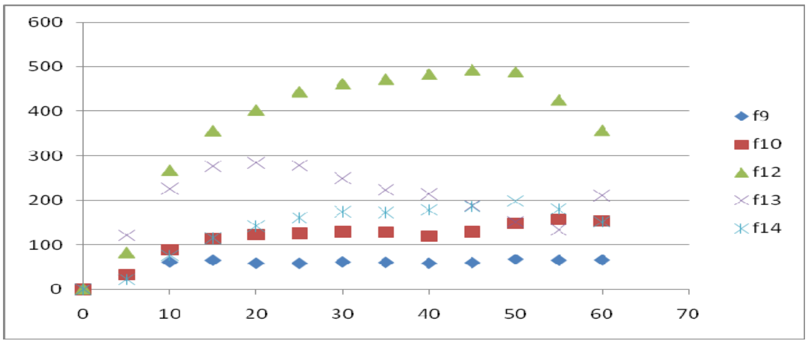


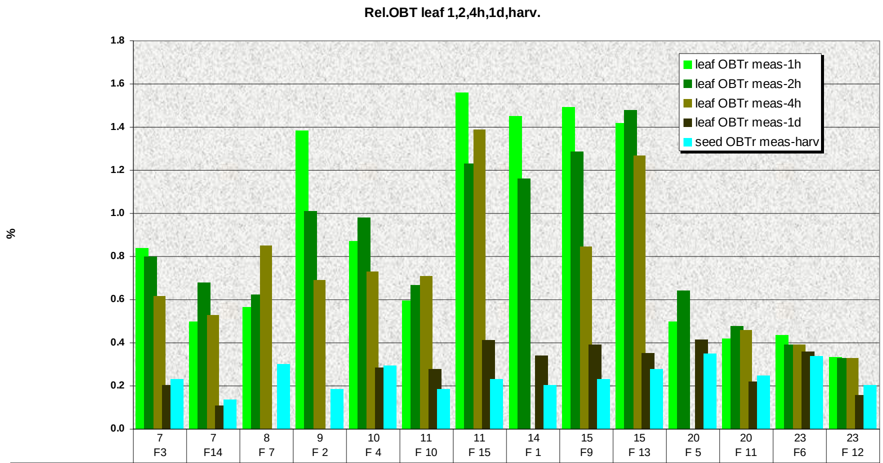
Figure G.III., Time dependence of HTO concentration in the box air moisture

The half lives of HTO in the first hour after the end of exposures are given in Table G.IV and demonstrate the diurnal pattern of canopy conductance

Table G.IV. Half-Lives of TWT concentration in wheat within 1 h after the end of exposure to HTO.

	TWT half-lives (min)			
Plant parts	Exposure at dawn (3 exp.)	Exposure at day-time (6 exp.)	Exposure at dusk (2 exp.)	Exposure at night (2 exp.)
Leaves	40-60	25-49	230-660	110-170
Stems	45-49	20-26	130-320	60-190
Ears	79-91	50-126	210-330	150-920
Total plant	50-72	27-60	220-340	100-250

If we take as a reference the HTO concentration in leaves at the end of 1 hour exposure, in order to have some normalization, the dynamics of OBT (combustion water) in leaves reveals interesting development (see Figure G.IV, taken from [17]). Immediately after the end of exposure, the highest relative OBT concentrations were observed in leaves under day-time conditions (1.25±0.34 %), about 3 times higher than under night conditions (0.38±0.05 %). In day time there is a clear reduction in the first day, due to assimilate export, which seems to start immediately after the end of exposure. In night conditions, the assimilate export is slower and perhaps more active in the next morning. Despite the large difference in leaf OBT at the end of exposure, in all experiments the OBT in grain at harvest shows similar relative values (mean = 0.25 ± 0.07 %). This can be partly explained by the longer residence of leaf HTO in night time (experiments F6, F12) allowing a larger contribution of metabolic processes to OBT formation. The translocation index for each experiment is given in Figure G.V.



	7 F3	7 F14	8 F 7	9 F 2	10 F 4	11 F 10	11 F 15	14 F 1	15 F9	15 F 13	20 F 5	20 F 11	23 F6	23 F 12
leaf OBTr meas-1h	0.8398742	0.4974324	0.5642564	1.3862011	0.8713017	0.597344	1.56	1.4502809	1.4935269	1.4188636	0.4996208	0.4180299	0.4357019	0.332595
leaf OBTr meas-2h	0.8014334	0.6785377	0.621814	1.0103351	0.9812237	0.6648128	1.23	1.160563	1.2869737	1.48	0.6446733	0.476718	0.391995	0.3312465
leaf OBTr meas-4h	0.6152852	0.5313257	0.8526928	0.6895166	0.73	0.71	1.39		0.8462179	1.2686161		0.4586435	0.391995	0.3312465
leaf OBTr meas-1d	0.2022103	0.1115011			0.2847011	0.278103	0.4105411	0.3408383	0.3936255	0.354457	0.4162202	0.2180439	0.3588892	0.1587702
seed OBTr meas-harv	0.2329832	0.1376657	0.3038118	0.1860488	0.2931243	0.1860569	0.2318863	0.2034843	0.2296406	0.2800298	0.35	0.245797	0.3387211	0.2045089

Figure G.IV. The dynamics of OBT in leaves (taken from [17])

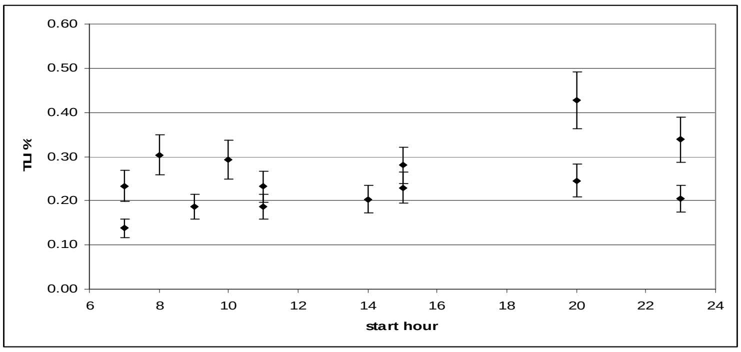
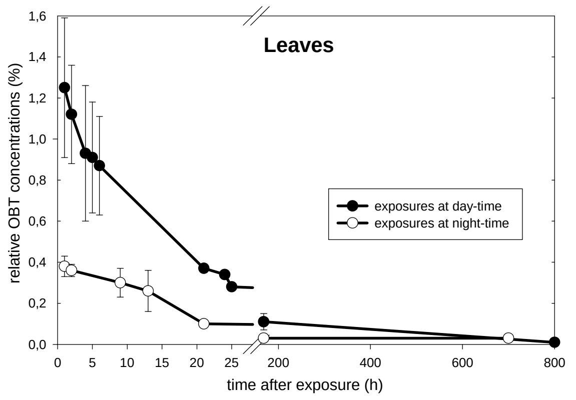


Figure G.V. Translocation index for wheat at different starting hours of exposure

The average dynamics of OBT in leaves and grain is given in Figure G.VI (taken from [19]) separately for day and night experiments. It seems that translocation in the night experiments is delayed until the next morning and is longer.



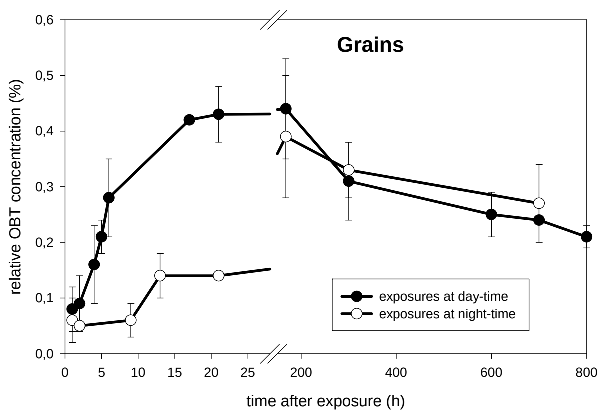


Figure G.VI. Patterns of relative OBT concentrations in leaves and grains between from exposure to HTO and harvest. The data represent means ± 1SD of 7 exposures under day-time conditions and of 2 exposures under night-time conditions.

The total OBT per plant increases in the first 2 days and can decrease until harvest at 80 % from maximum value. One example is given in Figure G.VII . The large decrease in OBT concentration in leaves is due to translocation in grain and to growth dilution. Maintenance respiration is low in grains, but explains the total plant OBT decrease.

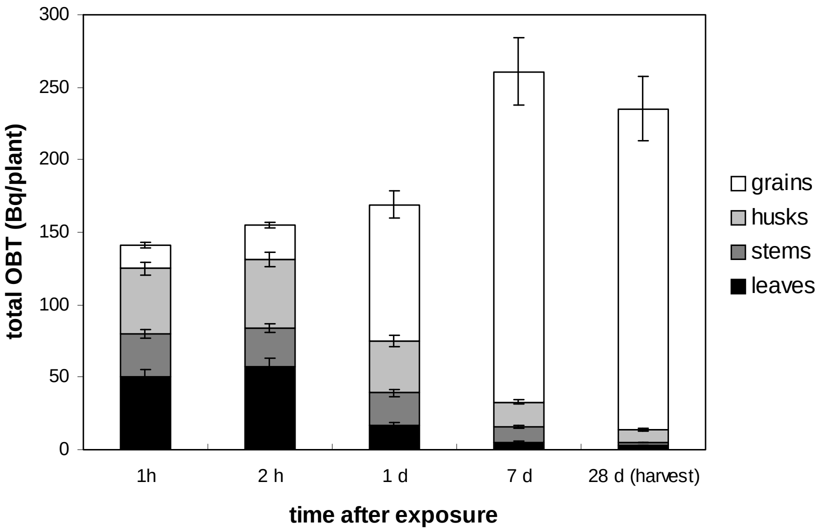


Figure G.VII. The distribution of OBT within the wheat plants exposed to atmospheric HTO during the dusk on the 20th day after anthesis (error bars represent counting error plus analytical error) (taken from [19])

An attempt to correlate the OBT in grain with the integrated HTO concentration in leaves and ears has been tested [19]. The best fit was obtained adding to the leaf concentration a contribution of ears with a 0.5 factor and using a general factor of 0.2 for the night integrated concentration (see Figure G.VIII). From this figure
OBT (Bq/kg) = 0.6 * 0.48* INT (kBq hour /L) = 0.6 * 0.48 * INT (Bq_d/l) * 24/1000 = INT (Bq d/L) * 0.27/40
This results show a simple relation between OBT concentration in grains and the integrated concentration in leaves and ears. Up to now, there is no clear explanation if this correlation is real.
Concerning the relative high OBT concentration in the grains following an exposure at dawn and during the night, when the plants were exposed in partial and complete darkness, respectively, the insights gained from the chamber experiments could be confirmed. The gas exchange measurements pointed out that the assimilation in the chloroplasts starts early in the morning when the TWT concentration in the leaves is still clearly enhanced [23].
For wheat, the experimental data show that translocation factor in night is close with the day value, based on chamber and field experiments. For 1 h exposure the average value is 0.23 %.

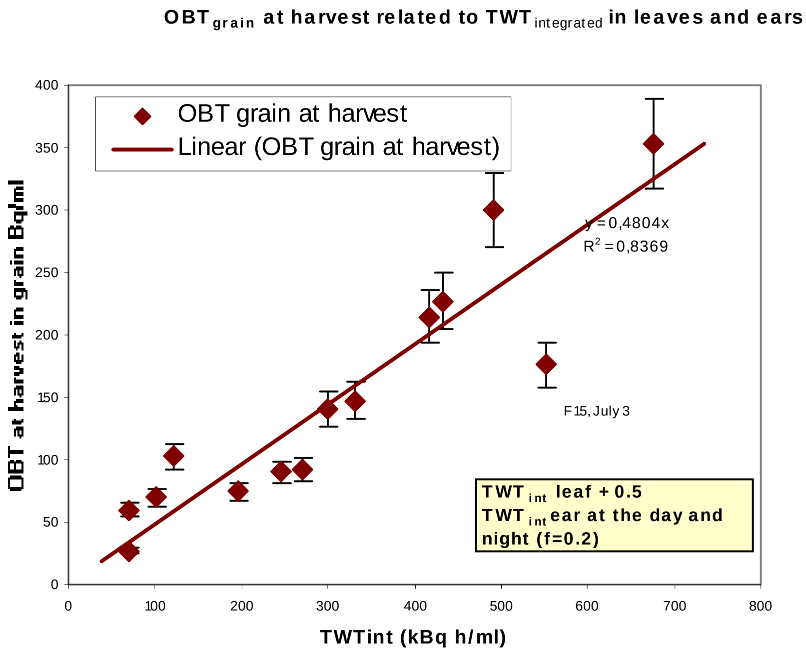


Figure G.VIII. Correlation between OBT in grains and integrated HTO concentration in leaves and ears (taken from [19])

A limited number of data with chamber experiments were obtained for **bean and potato** [24]. Cultivar used for bean (*Phaseolus vulgaris L*., cv. Hilds Maja) and potato (*Solanum tuberosum L*. cv. Erstling si Sieglinde) were grown in pots and the experimental protocol was the same as for the wheat in the first experiment described [19]. The experiments were done at 0-24 days after flowering for the bean and 13-45 days for potato. After 2 hours of exposure in the night, the relative concentration in leaves was 15 % in bean and 14 % in potato, while in the light (900 μmol m^{-2} s^{-1}) were 77 % and 83 %. This data are close with the wheat case and demonstrate a relatively high leaf conductance in night.
The relative OBT concentration at harvest (in terms of leaf HTO at end of 2 hours exposure) is given in Table G.V.

Table G.V. Experimental data for bean and potato

Plant part	Exposure at 900 µmol m^{-2} s^{-1}	Exposure at 120 µmol m^{-2} s^{-1}	Exposure at night
Beans			
Time after flowering	4	1	12
Leaves	0.7	0.3	0.5
Stems	0.4	0.2	0.3
Pods	0.1	0.03	0.4
Potato			
Time after flowering	20	13-25	15-23
Leaves	0.2	0.2	0.2
Stems	0.2	0.1	0.2
Tubers	0.3	0.2	0.2

As a first impression, it seems that night translocation in beans is close with wheat, but lower for potato. More experimental data are needed.

During the night exposures, the rate of OBT formation was 4-5 times lower than under day-time conditions because the photosynthetic system was not active. OBT has been generated by metabolic processes independent of light which are responsible for energy supply, growth and maintenance of plant structures.
As it was already shown in the above studies with spring wheat, green beans and potatoes, the storage organs represent a sink for organically bound tritium within agricultural ecosystems, which should be considered in models predicting the dynamics of tritium transfer in these systems

III. JAPAN: RICE, SOYBEAN AND TANGERINE

Due to restrictions on radiation protection, Japan has used deuterium in a series of experiments. Deuterium water (D_2O) release experiments were conducted using a greenhouse at Mito site of Ibaraki University for many kinds of vegetations or a climate control chamber in a laboratory of Japan Atomic Energy Agency for rice to simulate an environmental release of tritium from a nuclear facility [25, 26, 27, 28, 29, 30].

Rice

Rice is an important food item in Japan and in many Asian countries and contributes up to 36 % to the intake of background tritium. Potted rice plants were exposed to D_2O vapor in two identical greenhouses [7.2 m(L) × 1.8 m(W) × 1.8 m(H)] used separately for daytime and nighttime experiments [25]. The ripening period of rice is the stage between flowering (anthesis) and harvest and is 40–60 days depending on cultivar and climatic conditions. A rice cultivar (Yumehitachi) harvested after 25 days on was used in the experiment. Rice plants were grown in both flooded and non-flooded pots. D_2O vapor, with a concentration of 20 %, was released using an ultrasonic humidifier in a small box connected to the greenhouse and supplied with a flow of dried air. The inside temperature was 29.6–34.1°C during the day and 19.6–23.6 °C during the night. The air relative humidity was 44–57 % in the daytime and 71–94 % in night time. Light intensity, as photosynthetic photon flux density, was 158 µmol $s^{-1}m^{-2}$ on average during 8 h of daylight range (23–370 µmol s^{-1} m^{-2}). The release duration was 8 hours and the deuterium concentration increased gradually. Deuterium concentration in air moisture in the daytime did not increase as they expected, so a larger amount of deuterium water was released during the nighttime experiment. Therefore, deuterium concentration in air moisture was much higher than that in the daytime.

The concentration of the tissue free water deuterium (TFWD) in leaves followed the air D_2O concentration, and the ratio reached 0.53 at the end of exposure. The rate constants of D_2O uptake in leaves in the daytime were 4–5 times higher than that in nighttime. At the end of release the organically bound deuterium (OBD) in leaves normalized to the same air concentration was 3–4 times less in nighttime than in the daytime, and in grain the difference was greater. After the exposure stopped, the OBD concentration in grain under daytime exposure continued to increase for about 4 days, but for nighttime exposure the duration was longer. Then due to growth dilution, the OBD concentration in grain decreased until harvest. The translocation indexes referring to deuterium concentration in air moisture at the end of the exposure (TLIa) were near 0.2 % for unhulled rice and near 0.3 % for hulled rice for both the daytime and nighttime experiments, but that for day was slightly higher than that for night, for both flooded and non-flooded cases. The translocation indexes referring to leaf TFWD at the end of the exposure (TLI) were 0.42 % for day unhulled, 0.73 % for day hulled, 0.36 % for night unhulled and 0.54 % for night hulled. Considering the release duration of 8 hours, this pattern is lower than in the case of wheat (0.23 % for one hour exposure).

In the same time and same greenhouse, a different rice cultivar (Koshihikari) at 1 week after flowering was exposed to D_2O vapor [27]. The background deuterium in Mito air was 150 ppm (D/H) and that in OBT of grain was 144 ppm. After 8 hours of exposure, OBD concentration in rice grain increased with 25 ppm, both during the day and night.

The day/night ratios of OBD concentration normalized to the same air concentration were 2.5 for the grain and 2.9 for the leaf. OBD concentration in grain was investigated by the harvest on the 45th day after the experiment, and the OBT concentration in grain after the nighttime exposure decreased faster than that after daytime exposure. The TLI (referred to TFWD at the end of exposure) was 0.36 % for the day case and only 0.03 % for the night case. For this case with rice at the start of grain filling, the night translocation index is 10 times less than that for the day. The time dynamics of TLI until harvest show a clear distinction between day and night (see Figure J.I).

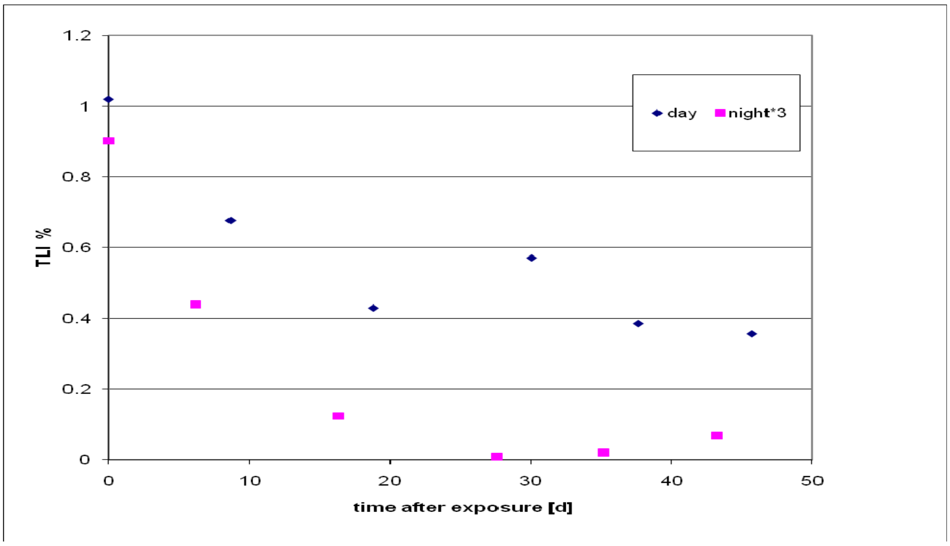


Figure J .I. Time dynamics of translocation index as function of time after exposure

Potted rice plants were exposed to D_2O vapor in a climate control chamber, as a substitute for tritium, for 4 hours at five different times during the grain-ripening period, in order to estimate the influence of the growth stage on the formation and retention of OBD in rice [28]. The plants were grown outside before and after the exposure experiments and were exposed to D_2O vapor in a laboratory in a small chamber equipped with controllers of temperature, humidity and light intensity. Concentrations of TFWD and OBD in rice leaves, stems and grains were investigated up to the harvest time.

At the end of the 4-hours exposure, TFWD in leaves reached 55 – 74 % of the D_2O concentration in air moisture. After one day the TFWD in leaves was close to the background concentrations. The TFWD in the grains remained for a longer time after the end of exposure than in the leaves and stems. The mass of OBD in grain at harvest showed the highest value when the exposure was conducted in the early stage of the ripening period. When the exposure was conducted after 26 days from the flowering, the increase of OBD in the grain was small. Most OBD in rice was formed within 24 hours from the initiation of exposure and transferred to the grain immediately. The mass of OBD in grains did not decrease until harvest. The translocation indexes for grains and stems are given in the Table J.I for each experiment performed at DAF (days after flowering).

Table J.I. Translocation index for rice (4 hour exposure)

DAF	6	14	21	26	35
TLI (%) grain	0.39	0.55	0.43	0.15	0.045
TLI (%) stem	0.05	0.03	0.04	0.03	0.03
TLI (%) leaf	0.19	0.14	0.17	0.18	0.12

Soybean

A preliminary experiment with soybean [24] at an early stage of pod development shows the TLI at a low value (0.14 % for day and 0.17 % for night, when referring to TFWD).

More D_2O release experiments were conducted for soybean [29]. Soybean was grown on the ground in the greenhouse. Exposures were performed at various stages of soybean development for 8 hours and harvest was for relatively young and fully ripened beans (consumption of green soybeans is common in Japan). The age dependence of the uptake rate as TFWD was observed and, in many cases, the night uptake rate was close to the day rate.
1999: exposure Aug 21, 22; harvest Oct 23
2000: exposure Aug 10 and Sept 12; seeding June 6, flowering July 16, harvest Oct 10
2002: exposure Aug 27 and Sept 12; A: flowering Aug 14, B: flowering Aug 18

In Fig. 3 in [29] (given below), the translocation indexes for young beans (G) and ripened beans (Y) are given for 2000 and 2002. For the exposure in 2000, the beans exposed on August 10 were in the early middle stage of growth and on 12 September in the late stage. When in the early stages, TLI was near 0.2 % in day exposure and lower at night. For 2002 exposure, soybean on August 27 was in the early stage of growth (Fig. 3 in [29]) and on September 12 was in the early retardation phase (Fig. 4 in [29] anf given below). The highest TLI of 4.3 % (day) was for a few days after exposure, but at harvest decreased to 0.8 %. TLI for green beans in night was 1/5 from those in the day, while for ripened beans the ratio was 1/2. Table 4 in [29] (given below) summarizes the soybean results (referring to air concentration) and demonstrates that TLIa during the night is lower than that during the day. If we consider the definition of TLI in relation to leaf water, from Figs. 3 and 4 we can observe that soon after exposure, TLI is high and decreases until the final harvest (28 days later). At final harvest, TLI is near 0.7 % for the daytime experiment and 0.5 % for night. In the reference [29],, the details on the pod development stage at exposure are difficult to assess. More information is required but the researcher retired.

Note that these results are for an exposure of 8 hours, when air concentration gradually increased in the glasshouse. A rough translation for one-hour exposure is to divide the TLI by 4–6.

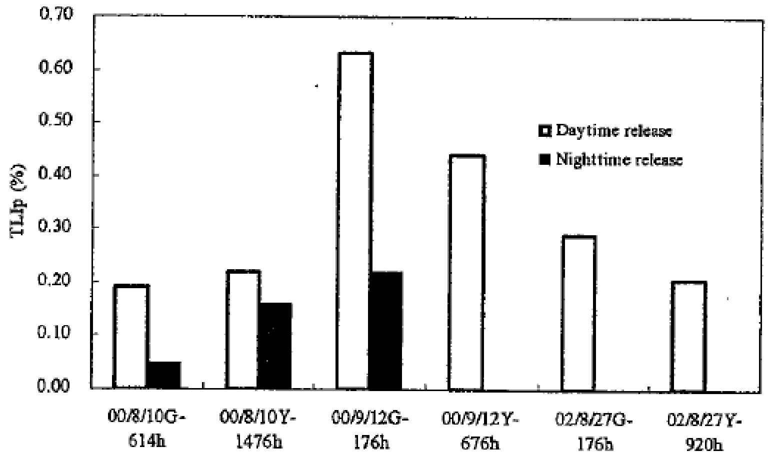


Fig.3 Translocation index (%) in young (G) and ripend (Y) soybean in 2000 (August and September) and 2002 (August)

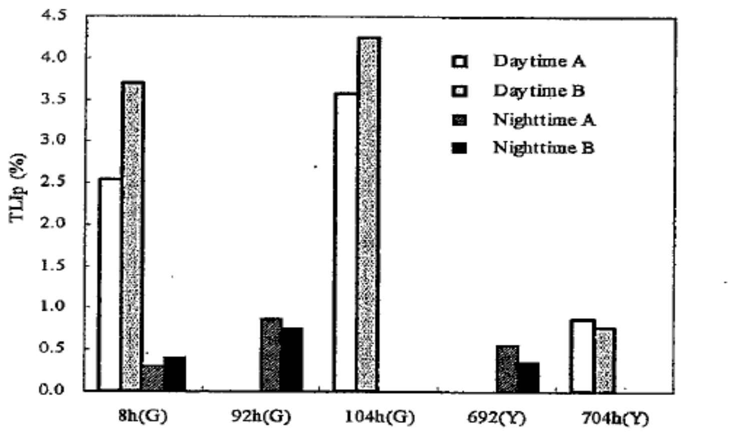


Fig. 4 TLI p (%) in young(G) and ripened (Y) soybean harvested at 8, 92, 104, 692 and 704 hours after the start of D2O exposure on 02/9/12

Table 4 Translocation index (TLIa) in young and ripened soybean at harvest

		TLIa (%)			
		Daytime release		Nighttime release	
Exp. No.	Exp. Date	Young bean	Ripened bean	Young bean	Ripened bean
I	Aug. 99		0.08		0.08
II	Aug. 00	0.11	0.13	0.04	0.15
III	Sept. 00	0.43	0.33	0.22	
IV	Apr. 02	0.00	0.00	0.00	0.00
V	Aug. 02	0.21	0.15		
VI	Sept. 02 (A)	2.87	0.70	1.00	0.62
VI	Sept. 02 (B)	3.41	0.63	0.88	0.39

Tangerine

Tangerine (*Citrus unshiu* Marc.) was exposed to D_2O vapor in greenhouses at 6 times of D_2O release experiments in 2000 - 2002 [30]. Exposures were before and after flowering, as well as in the middle or end of the ripening period. The duration of exposure was 8 hours during the daytime and nighttime. Depending on the growth stage, the temperature, light and humidity, the uptake rate constants from air to tangerine leaf in the daytime varied between 0.2 and 1.1 h^{-1}. In night, the rate constants were several times lower (0.03–0.12 h^{-1}). These values were lower than for rice and soybean used in the greenhouse experiments, because cuticle structure for tangerine leaves differs from rice or soybean. After the exposure the loss rate constants in leaf TFWD were 0.8–1.2 h^{-1} in the daytime and 0.03–0.2 h^{-1} in nighttime. Lower values were observed in winter than in summer. The translocation index (as defined in this report) varied between 0.08 %–0.28 % in the daytime and 0.21 %–0.71 % in the nighttime with the maximum in the middle of the ripening period. **Note that for this experimental data, the night OBT formation is higher than the day one.**

IV. SOUTHERN KOREA: RICE, SOYBEAN CABBAGE, RADISH

Rice

The experimental techniques developed in Germany for wheat were applied in Southern Korea for rice, the main cereal in Asia [31]. A rice cultivar (*Oryza Sativa L.* var, called Jangan-byeo) was grown in plastic pot and exposed for one hour to HTO, in a Plexiglas box for one hour, at various time of development stage. The exposure was in day, around 10 a.m. and the conditions were chosen to reproduce the real situation in Southern Korea. Nine exposures were done, before heading and until yellow ripe of rice (20 days before the final harvest). All the conditions in the exposure time were registered (dynamics of air HTO concentration, light, temperature, and humidity). At the end of exposures, the leaf water HTO was at least 60 % of the air moisture and practically 100 % in optimal light and temperature conditions. Ear concentration was 35-100 %. After exposure, in the first 5-6 hours, the TFWT concentration was reduced by a factor of about 1000 in leaves and about 100 in steams. The decrease is much more rapid than for grape, potato and cabbage. The reduction factors (TFWT at the end of exposure/ at harvest) is higher than for tomato, potato, sunflower and maize. OBT in different plant parts at the end of exposure is influenced by the exposure conditions, but also by the plant development stage. The initial OBT content (water of combustion) at the end of exposure in leaves was 1.0-1.2 % from the TFWT at the end of exposure, comparable with the wheat data. The decrease in leaf OBT is slower than for leaf TFWT. At 4-5 hours post exposure, OBT predominates in leaves. Because wheat is used in human consumption as grains only, we are interested in the harvest value of OBT. Table K.I summarize the results for exposures at various time (DAA) related to flowering (booting). The relative stage refers to time between booting and harvest. The initial data were normalized to 10^4 Bq/ml of the initial leaf TFWT concentration. Yields of grain per shoot were 1.65 g.

Table K.I. Results for the Korean experiment with rice

Exp.	Date	Stage	DAA	Stage rel.	OBT grain per shoot	OBT grain (Bq/g)	OBT grain (Bq/ml)	TLI (%)
E1	11-Aug	bootig	-7	-0.130	0.480	0.291	0.485	0.005
E2	18-Aug	heading	0	0.000	0.930	0.564	0.939	0.009
E3	21-Aug	milky ripe	4	0.074	4.190	2.539	4.232	0.042
E4	25-Aug	milky ripe	8	0.148	8.530	5.170	8.616	0.086
E5	28-Aug	milky ripe	11	0.204	32.100	19.455	32.424	0.324
E6	1-Sep	dought ripe	14	0.259	36.300	22.000	36.667	0.367
E7	5-Sep	dought ripe	19	0.352	22.700	13.758	22.929	0.229
E8	10-Sep	dought ripe	24	0.444	18.300	11.091	18.485	0.185
E9	19-Sep	yellow ripe	33	0.611	3.170	1.921	3.202	0.032
Harvest	10-Oct		54					

Note that the experiments E1, E2, and E7 are less accurate than the rest of experiments, due to the experimental problems.

In conditions of full day, at the time of maximum TLI, the leaf TFWT at the end of exposure has the same concentration as the air moisture.

Night experiments were described in a different paper in Korean language [32]. One experiment was in the period of milky ripe and the other one in early dough ripe, 45 and 40 days before harvest (25 Aug and 1 Sept). Temperature and relative humidity were in the normal range (21 °C and 93 %, respectively). After one hour of exposure, the leaf HTO reached 30-40 % from the mean air moisture concentration. This is 2 times higher than for wheat and about 3 times lower than rice in full day. HTO concentration in ear reached 15-30 % from air moisture, again higher than for wheat. For leaves, OBT concentration in combustion water, at the end of exposure was 0.1-0.2 % from the concentration in leaf water. At harvest the translocation index for grain was 0.1-0.14 %, equal or slightly less than for full day. The authors appreciate that in night OBT was produced at about a third from the rate for full day (mostly due to lower uptake of HTO in night).

Soybean

In EMRAS I, it was a blind test of participated models, based on unpublished data on HTO exposure for a Korean cultivar of soybean [33]. The soybean scenario [33] addresses the tritium absorption by soybean foliage and the subsequent tritium behavior 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 of soybeans' growth. The tritium behavior in the plant body and pods was observed by sampling the various plant parts and determining their concentrations.

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 dates on which the exposures were made for SB1 to SB6 were July 2, July 13, July 30, August 9, August 24 and September 17, 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 usually, 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. The observed concentrations corresponding to the requested predictions are shown in Tables K.II, K.III and K.IV 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.

Table K.II. Observed and normalized tissue free water tritium concentrations (TFWT) for SB1 experiment

In the plant body (stem and leaves)			
Time and date	**Elapsed time after exposure (hr)**	**TFWT (Bq/mL)**	**Normalized TFWT***
10:40 July 2	0.2	9580	$1.23x10^{-1}$
11:30 July 2	1	1050	$1.35x10^{-2}$
July 3	24	3.92	$5.05x10^{-5}$
July 7	120	1.32	$1.70x10^{-5}$
July 16	336	0.33	$4.25x10^{-6}$
Aug. 10	936	0.11	$1.42x10^{-6}$
Sept. 7	1608	0.06	$7.73x10^{-7}$
Oct. 5	2280	0.06	$7.73x10^{-7}$
In the pods (shell and seeds)			
Time and date	**Elapsed time after exposure (hr)**	**TFWT (Bq/mL)**	**Normalized TFWT***
Aug. 10	936	0.21	$2.70x10^{-6}$
Sept. 7	1608	0.06	$7.73x10^{-7}$
Oct. 5	2280	0.06	$7.73x10^{-7}$

* Tissue free water tritium concentration in the plant divided by the average tritium concentration in air moisture during the exposure (7.77 x 104 Bq/mL for SB1).

Table KIII. Observed and normalized TFWT concentrations for SB4 experiment

In the plant body (stem and leaves)			
Time and date	**Elapsed time after exposure (hr)**	**TFWT (Bq/mL)**	**Normalized TFWT***
10:40 Aug. 9	0.2	7000	$1.33x10^{-1}$
11:30 Aug. 9	1	3200	$6.08x10^{-2}$
Aug. 10	24	25.9	$4.92x10^{-4}$
Aug. 14	120	2.1	$3.99x10^{-5}$
Aug. 23	336	0.8	$1.52x10^{-5}$
Sept. 10	768	0.27	$5.13x10^{-6}$
Oct. 5	1368	0.14	$2.66x10^{-6}$
In the pods (shell and seeds)			
Time and date	**Elapsed time after exposure (hr)**	**TFWT (Bq/mL)**	**Normalized TFWT***
10:40 Aug. 9	0.2	10500	$1.99x10^{-1}$
11:30 Aug. 9	1	8000	$1.52x10^{-1}$
Aug. 10	24	2700	$5.13x10^{-2}$
Aug. 14	120	63.5	$1.21x10^{-3}$
Aug. 23	336	1.49	$2.83x10^{-5}$

Sept. 10	768	0.84	$1.59\text{x}10^{-5}$
Oct. 5	1368	0.26	$4.94\text{x}10^{-6}$

* 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.

Table K.IV. 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	2.06×10^{-4}	0.83	0.5	0.67	8.63×10^{-6}
SB2	1.47×10^{5}	59.8	50.8	55.3	3.75×10^{-4}	3.5	3.7	3.6	2.44×10^{-5}5
SB3	1.14×10^{5}	37.8	17.7	27.8	2.44×10^{-4}	101.3	19.3	60.3	5.28×10^{-4}4
SB4	5.27×10^{4}	19.8	8.8	14.3	2.71×10^{-4}	74.7	200.0	137.4	2.61×10^{-3}3
SB5	9.19×10^{4}	44.3	13.5	28.9	3.14×10^{-4}	73.3	214.2	143.8	1.56×10^{-3}3
SB6	1.37×10^{5}	180	19.5	99.8	7.28×10^{-4}	33.5	77.0	55.2	4.03×10^{-4}4

[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

For experiments SB3 through SB6, the OBT 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.

The observations clearly show that the OBT concentrations in the pods relate to the stage of growth at which the exposure occurred. The OBT concentration increased with experiments SB1 through SB4, which covered the period of active plant growth. The normalized OBT concentrations in the pods for SB3, SB4 and SB5 were much higher than in the leaves. The exposures for these experiments were made when the fruit were developing rapidly and most tritium absorbed through the leaves may have been transferred directly to the pods rather than being stored in the leaves. The relatively low concentration in the pods for SB6 may be due to the fact that development of the pods was close to complete when the exposure occurred. This might cause a reduction in the tritium transfer to the pods and in the OBT concentration in them.

The experimental data includes leaf HTO at the end of exposure only, for SB1 and SB4 and relative to the air moisture concentration the values are low about 0.13, showing a low uptake rate (lowest than for the Japanese experiments). Adopting the same relative uptake for all experiments, a translocation factor (as it was defined in this report) can be obtained (see Table K.V).

Table K.V Approximate translocation index for Korean soybean

Case	**DAA**	**TLI (%)**
SB1	-5	0.005
SB2	6	0.019
SB3	23	0.13
SB4	33	2.9
SB5	48	1.8
Sb6	60	0.42
harvest	78	NA

If we compare this experimental data with the Japanese ones, we observe a higher maximum TLI and this remains to be checked. An explanation can be an incomplete extraction of exchangeable OBT.

Radish

Radish was used as a typical root vegetable in many experiments, including the long release of HT in Canada (1994). A Korean cultivar (*Raphanus sativus L.*) was used for short exposure (1 hour) at various stages of plant development [34]. Contrary to cereals, radish has large water content in leaves (92 %) and root (94 %). The exposure conditions, as well as plant growth were known and the ratio between fresh and dry weight for top of the plant and root is known, also. At the end of exposure, the TFWT (tissue free water tritium) at the top was 1-2 orders of magnitude higher than for roots, but later at harvest, they were comparable. When the plants are exposed to tritium near thei the harvest time, the concentrations in tops and roots at harvest are increased, as it was expected. The edible plant part of radish is root only and we are interested in tritium contamination of the root. Constrained by the atmospheric forces, HTO moves to root opposite to the main water path (root to leaves). Most of HTO translocated to root is transported within assimilate (phloem) and not counter-gradient within the water in xylem. Relative to mean air water concentration in the exposure time, the root TWT at harvest is minor and is higher than 0.001 (relative units) only 2 weeks before harvest.

From tables and figures in the cited reference [34], we can get some important results. At the end of exposure, leaves HTO concentration is 20 - 40 % from the mean concentration in air moisture, a smaller value than for cereals. The root OBT at harvest (Bq/g dm) is no more than 0.3 % from the concentration of HTO in the leaves water at the end of exposure. At harvest, the ratio of OBT to TFWT concentration strongly depends on the exposure time before harvest and decreases when the exposure takes place near the harvest time. Excepting the exposure close to harvest (<7 days), the contribution of OBT to the ingestion dose is the highest. The translocation index, as it was defined, is less than 0.4 %. Table K.VI summarize the experimental results. DTH indicate day to harvest; Cend is the HTO concentration in radish leaves at the end of exposure while rel. Cend refers to the ratio of HTO concentration in leaf water at end exposure and the averahe HTO concwentration in air moisture in the exposure time. For radish root at harvest, the OBT concentration (Cobt_root) is given in the Table K.VI, as well as the translocation index.

Table K.VI. Summary results for the tritium contamination (1 hour exposure) for Korean radish

DTH	45	35	27	15	7
Cend (kBq/mL)	21	25	40	37	42
rel Cend (%)	46	40	22	33	44
Cobt_root (Bq/mL, OBT water of combustion)	23	75	113	93	140
TLI (%)	0.11	0.30	0.28	0.25	0.33

Cabbage

In the same reference [34], there are reported the results for a leafy vegetable. A Korean cultivar of Chinese cabbage (*Brassica pekinensis Rupr.*) was exposed various times for one hour in a box with known HTO vapor concentration, temperature, humidity and light. The top of cabbage contains 95 % water and the leaves number, as well as the fresh mass increased significantly from the first exposure (51 days before harvest) until harvest. The earlier appearing leaves of Chinese cabbage become the outer leaves as time goes on. Around the end of the first half of the growing period, the leaves begin to erect themselves to form a tight leaf arrangement. During the late growth stage, several old leaves become senescent and fall off the plant. In this case, the outmost leaf of the inner part takes the place of a falling leaf to join the outer part. Outer leaves are more exposed to contamination than inner ones and the canopy structure changes in the time of plant development. A summary of exposure conditions, concentration of HTO in tops at the end of exposure and the concentration of OBT at harvest is given in Table K.VII, as well as the translocation index

Table K.VII. Experimental data for Korean cultivar of Chinese cabbage

experiment	**C1**	**C2**	**C3**	**C4**
DTH	51	42	29	17
leaves number	18	22	30	40
fresh mass	170	500	850	1000
start hour	10	10	10	10
T (°C)	26.8	29.1	30.4	19.4
RH (%)	86	83	78	89
light (klux)	42	52	61	23
mean HTO (kBq/ml)	71	30	43	147
Cend (kBq/mL)	34	18.8	12.3	14
rel. Cend (%)	48.2	62.2	28.4	9.5

Cobt_top (Bq/mL, OBT water of combustion)	17	32	56	105
TLI (%)	0.05	0.17	0.45	0.75

The generally higher relative OBT concentrations in the present experiment are partly attributable to the fact that the leaves of Chinese cabbage are not only the site of OBT production but also the main site of OBT storage.

V. CANADA: CHERRY TOMATO

The ETMOD model used in Canada by AECL has not considered the OBT formation in plants during the night. A study on relevant processes was done [35] and a conceptual model was proposed [36]. In order to clarify the aspects of OBT formation in the night, an experimental study to determine the transfer rates of tritium from air to the leaves and fruits of tomato plants following acute HTO exposure was done [37, 38]. Through a series of one-hour experiments carried out both during the night and during the day, a dataset on the build-up of OBT concentrations in the plant leaves and fruit was collected. The experiments were carried out using an exposure chamber with cherry tomato plants at three different growth stages (early, intermediate, and late). Separate experiments were conducted to determine the short-term and long-term dynamics of HTO and OBT concentrations in the leaves and fruits from the end of exposure to harvest. Both HTO and OBT concentrations in leaves at the end of the exposure were greater in the experiments carried out during the day than those carried out at night. This supports the belief that OBT is produced mainly by photosynthesis in the presence of sunlight. OBT can be produced in leaves in the dark, but through other processes that operate at slower rates. At harvest, the HTO and OBT concentrations in the fruit depend on the length of time between exposure and harvest and on the time of day at which the exposure was carried out. HTO persisted much longer in the fruit than in the leaves, but OBT showed a similar persistence in both plant parts.

Cherry tomatoes (*Lycopersicon esculentum* var. cerasiforme) were used in this study. Experiments started at the time of flowering and included a period of small fruits formatting, as well as a period with ripened and red fruits. Day experiments included only the ripened fruit period. Leaf area per plant, leaf water content, fruit growth and starch dynamics in the leaves were measured. Transplanting was done on June 3, 2002, when the plants were about 3 weeks old, and the final harvest was in late September. The growing season was longer in the field due to the climate in Chalk River. Experiments 1 to 6 were done in the dark and experiments 7 to 8 were done in the daylight. If the definition of the translocation index is accepted (harvest OBT/leaf HTO at the end of exposure), the tomato data reveals interesting results (Table C.I).

Table C.I. Translocation Index results for cherry tomatoes

Experiment	Day after exposure	TLI (%)	Remarks
Exp 1	90	0.00356	Night exposure, no fruit
Exp 2	4.3	2.12	Night exposure, no fruit
Exp 3	74	0.016	Night exposure, green fruit
Exp 4	0.58	0.149	Night exposure, green fruit
Exp 5	42	0.51	Night exposure, ripe fruit
Exp 6	1.7	0.346	Night exposure, ripe fruit
Exp 7	49	0.44	Day exposure, ripe fruit
Exp 8	48	0.12	Day exposure, ripe fruit

In experiment 2, at 7 days after flowering, there was an abnormally high translocation index. In experiment 4, the fruit was measured 14 hours post-exposure, and, if growth dilution is considered, the translocation factor is close to 0.007 %, only two times lower than in experiment 3. Experiments 5 and 6, carried out during the night with ripened fruit, showed consistent values at about twice the nighttime values from cases in Germany. Unexpectedly, the day cases in experiments 7 and 8 showed lower values than the night case. The fact that the value for a cloudy day was higher than for a sunny day was also unexpected. Ripened fruit may not influence the OBT produced in leaves. After analyzing the details of the experimental protocols, the causes for this have not been identified. The huge value (2 %) in experiment 2 is also a mystery. The observation that day translocation is lower than the value during the night when fruit is ripe deserves more investigation.

Because the OBT in fruits is translocated from leaves, it is interesting to compare the OBT dynamics in leaves during the day and night. The data were normalized to the HTO concentration at the end of exposure and Figure C.I shows only the values for ripened fruits. The period of the increased OBT concentration in the leaves was shorter (2 to 6 hours) in the daytime experiments than in the nighttime experiments (>2 hours). The decrease in OBT was faster during the day.

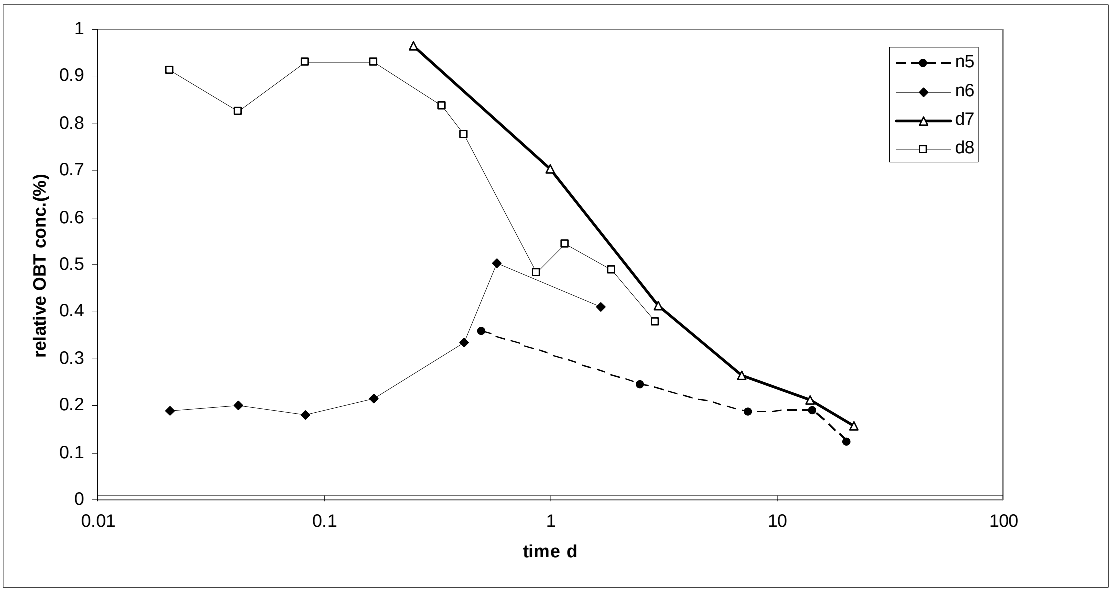


Figure C.I. Dynamics of relative OBT concentration in leaves

Comparing with the data for wheat (Figure G.VI, the values at the end of exposure are comparable and slightly lower.

When fruits were ripened (experiments 5-8), the initial uptake of HTO in the leaves was about 6 % during the day and 2.5 % during the night. For the immature fruit period, the night uptake was higher, closer to 5 %. All these values are definitely lower than for wheat case. The night canopy resistance is a plant-specific trait. The large leaf resistance during the night will maintain the HTO concentration until the next morning and the leaf OBT will contain a large part of what was produced during the day.

HTO and OBT trends for daytime and nighttime experiments were clearly different. However, the ingestion dose, which depends primarily on the OBT concentration in fruit at harvest, was more similar for daytime and nighttime exposures.

In order to understand the OBT production, more experiments were done. The daily starch production was measured (Figure C.II), as well as the starch pattern and values at dusk and dawn (Figure C.III and C.IV). The modeling trials up to now were not successful in explaining the experimental data in tomato fruits.

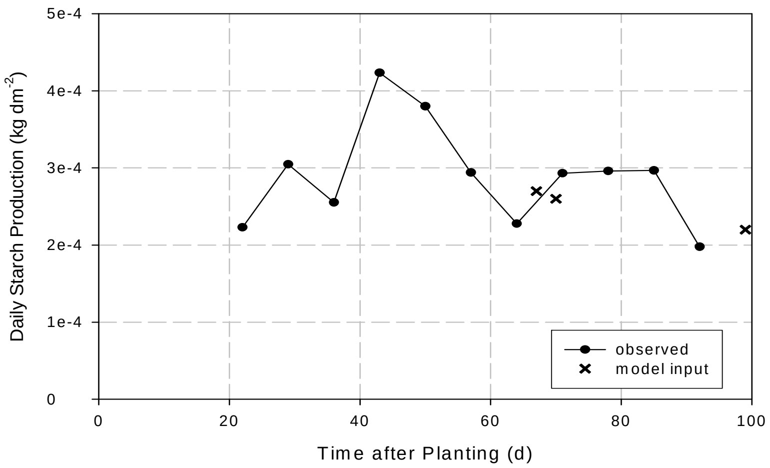


Figure C.II. Daily starch production per unit area of the tomato leaf

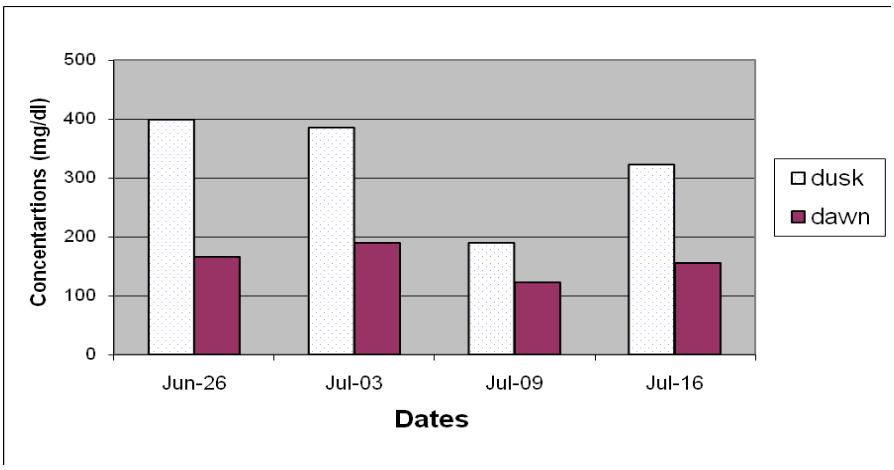


Figure C.III. Starch concentration at dusk and dawn in tomato leaves

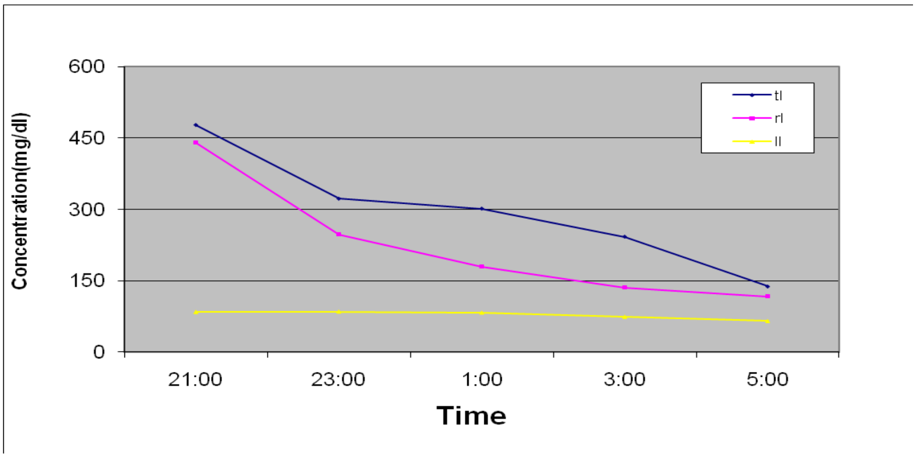


Figure C.IV. Pattern of starch concentration in night for tomato (tl), radish (rl) and lettuce (ll)

VI .FRANCE: LETTUCE

Experiments with lettuce (variety “feuille de chene blonde”, or *kitare*) were done in France recently [39] using a controlled climatic experimental chamber or in the field. For the chamber experiments we remark that the luminance used is lower than the normal ambient one and far from the saturation of photosynthetic rate. For tritium measurements, we note that two methods were used to extract the exchangeable form: the well known cold isotopic exchange, and a new one, based on a hot technique used in analytical chemistry. There is not a clear comparison between the two approaches and finally, there are data on only the total organically bound tritium. The method used for extraction of organically bound tritium was combustion and catalytic oxidation. The experimental errors are higher for samples with a mass less than 1 g. Daylight conditions or night conditions were maintained for 24 hours and the kinetic of HTO incorporation were followed for young, mature and relatively late mature plants (40, 60 and 70 days after germination). Temperature, humidity and light conditions were relatively close for all plant ages. The transfer of tritium from air to leaf is regulated by stomata resistance and we expect slower transfer in the night. There is a direct link between stomata resistance and photosynthetic rate, measured for many leaves and ages. Looking on the measured photosynthetic rates of leaves, we expect that the late mature plant (before flowering will have more old leaves but also higher surfaces. It is quite difficult to assess the canopy photosynthesis from the data presented, and consequently the exchange velocity. In the cited reference [39] and in our own approach, it remains to understand why the young plant shows no day/night difference.

Considering also all results presented in this report, we see that in the night the transfer rate is 3-5 times slower than in the day. It deserves more research because this aspect can have an important radiological impact.

In the controlled experiment the OBT/TFWT ratio is about 5 and not smaller than 1, as it was expected. All the potential experimental errors have been analyzed and it seems that luminance was too low comparing to outdoor. The experiments were done in a room in the Valduc waste treatment, a technological area. This can contaminate the air with many impurities These impurities can concentrate in the lettuce and later in the combustion water as volatile organic compounds. With simple purification of combustion water some significant error can occurs. For outdoor experiments the tritium is exhausted from the stack, after filtration and averaged on all the rooms in the facility. It can contain few impurities, if any.

Due to this potential cross contamination of OBT in the controlled experiments, the analysis of OBT production is misleading, but the controlled experiments are extremely useful for understanding the plant growth and uptake of HTO from air and soil into the leaves water (for model testing).

For the long term exposure under natural conditions (EXT1 and EXT2), the incorporation rate of OBT reveals very interesting results. First, in a static approach (as ratio of total OBT to integrated HTO concentration in the air moisture or plant water), the data are in the range of other experimental values – a mean transfer rate between 0.13- 0.16 (% h^{-1}).

The dynamic treatment of OBT incorporation demonstrates the variability of the process rate and the clear link with plant growth processes and environmental conditions. The first experiment was done from end of April to beginning of July with optimal conditions for temperature and light, while the second one was done in late autumn and the plant growth was slower. When the plant growth dynamics and OBT incorporation is considered in terms of available energy, it is seen that both cases have similar dynamics for the same total available energy. Also the OBT incorporation rate depends on plant development stage and environmental factors. The experimental results and the above conclusions can be used for testing the recent dynamic models for tritium.

VII. EXPOSURE TIME DEPENDENCE OF TLI

Only for the day exposure, there are experiments for rice - 1 hour (Choi etall [31]), 4 hours (Atarashi-Andoh et all [28]), and 8 hours (Ichimasaet all [26],Atarashi-Andoh et all [27])

We will refer to TLI as a function of time between the flowering and the final harvest. The stage is the exposure time related to the flowering and divided by the time between the flowering and the final harvest.

Rice - 8 hours exposure (Atarashi-Andoh et all [27])

Stage	TLI (%)
0.1	0.36

Rice - 8 hours exposure Ichimasa et all[26]

Stage	TLI %
0.45	0.63

Rice - 4 hours exposure (Atarashi-Andoh et all[28])

Stage	TLI (%)
0.14286	0.4
0.33333	0.55
0.5	0.42
0.61905	0.15
0.83333	0.045

Rice - 1 hour exposure (Choi et all[31])

Stage	TLI (%)
-0.0526	0.005
0.07018	0.0093
0.12281	0.04
0.19298	0.085
0.24561	0.32
0.31579	0.363
0.38596	0.227
0.47368	0.186
0.66667	0.032

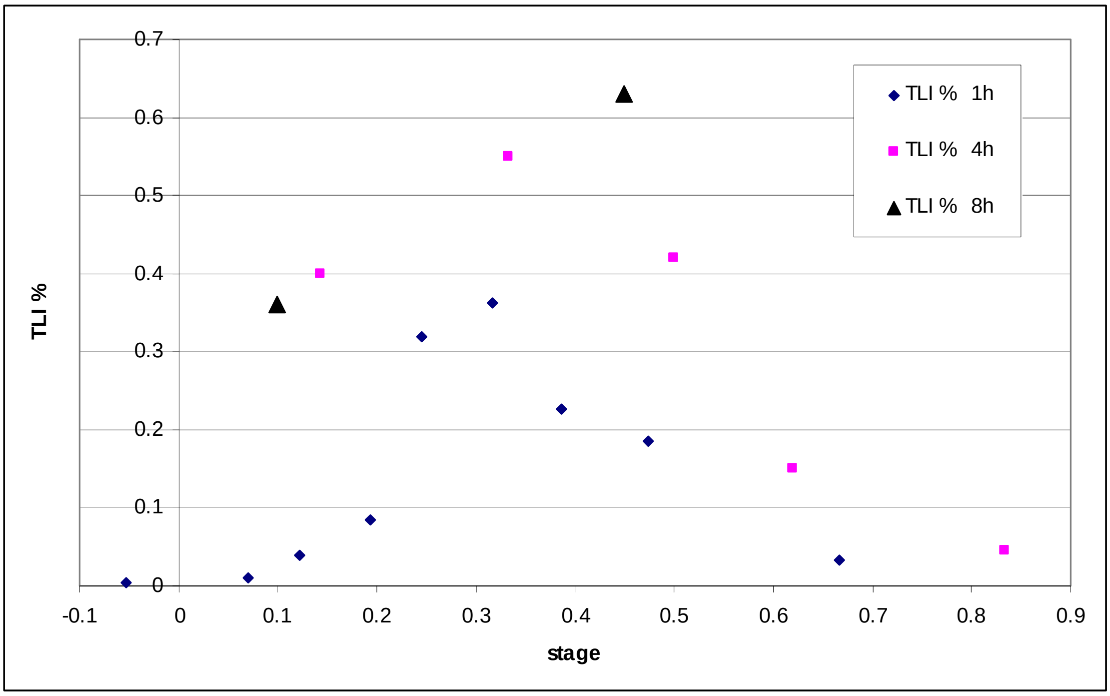


Figure VII.1. Comparison between different experimental data [26,27,28,31]

Comparing the data (Figure VII.1) there is clear that not only the duration of exposure influence the TLI at the same stage, but the cultivar characteristics, meteorological factors and details on HTO and climate evolution in the exposure time, also.

VIII. UPTAKE OF HTO IN NIGHT AND LEAF RESISTANCE

If the stomata are completely closed, it remains the cuticle resistance, only. Many models consider a default value about 4000 s/m. The cuticular permeability to water is usually characterized by the variable permeance (P), which is the ratio of water flow rate density to the driving force, the latter being expressed as a concentration difference [40]. At 25 °C and at standard pressure, the density of liquid water is 43 384 times greater than the saturation water vapour concentration in air and, correspondingly, the values of P referenced to the gas phase are greater by the same factor than values referenced to the liquid.

It is frequently observed the increase in cuticle permeability to water with the increasing air humidity and with temperature. The cuticle transpiration cannot automatically be assumed to be negligible. Aqueous pores enhance cuticular conductance [41]. The data on the water permeance in leaves [42] suggest a large variability of night uptake rate of water. In different tritium experiments, the low uptake has been seen for tomato leaves and lettuce [39] and high ones for sunflower. Rice has a high night uptake and wheat, bean and potato are at intermediate range. It will be useful to have direct experimental data for each major crop of interest. Atarashi et al. [43] measured stomata resistance in night with porometer and they found the following values: for komatsuma and radish - 300 s/m, but 700-1500 s/m for cherry tomato. Stomata resistance decreases 2 times when the relative humidity increases from 40 % to 90 %. Momoshima et al. [44] measured a value of 500 s/m for komatsuma, but only 250 s/m for cabbage.

When the data on deuterated water uptake was used [45] and compared with the porometer measurements, komatsuma shows variability between cases, cherry tomatos have about 2000 s/m and orange leaves had very high values (Table VIII.1).

In the experiment done in 1995 [45], komatsuna samples were at their young growth stage and the pot soils were not covered with a vinyl sheet. Therefore, the D_2O uptake by komatsuna leaves in 1995 seemed to include the uptake via root, which explains why the D_2O concentration would be higher.

Table VIII.1. Night uptake rate and resistance

Exp.	Plant	Rate (h^{-1})	Resistance using porometer (s/cm)	Rate using porometer (h^{-1})
Night 1995	**Komatsuna**	0.65 ± 0.19	5.7–40	0.06–0.44
	Orange	0.06 ± 0.29	49–55	0.04–0.05
Night 1996	**Komatsuna**	0.20 ± 0.04	2.7–3.2	0.82–0.97
	Radish	0.31 ± 0.05	2.6–3.4	0.72–0.95
	Tomato	0.12 ± 0.02	6.9–15	0.16–0.36

The exchange rates (h^{-1}) for lettuce [39] are very low (perhaps influenced by the low light) and shows the interesting fact that for the young plants, the day and night uptake rates are close and the mature plants (before flowering) have high resistances.

Table VIII.2. Lettuce uptake rates

Plant	Day	Night
Young	0.023	0.027
Mature	0.04	0.005
Before flowering	0.11	0.035

The experimental data up to now indicate that under quite identical condition plants differs in respect to stomata conductance.

IX. PRELIMINARY CONCLUSION FOLLOWING THE EXPERIMENTAL DATA OVERVIEW

We have find experimental data for wheat, rice, soybean, tomato, cabbage, radish, tangerine and preliminary results for lettuce and potato. Uptake and loss rates of HTO in plant water (leaves mostly) highly depends on environmental conditions (light, temperature and relative humidity), but also on crop characteristics such as stomata density and age of leaves. The abrupt changes of environmental parameters make the leaves to need some time to adapt to the new parameters, of about 10-20 minutes. Stomata resistance decreases by a factor 2 with increasing the relative humidity and decreases by a factor 2 - 4 with the leaf age [43]. The measured stomatal resistance has a diurnal cycle and seems to be different with each leaf of the same plant and same age [45]. Sunflower, grape, rice and wheat show higher uptake rates than komatsuma and cabbage, while tomato and orange have the lowest uptake rate [43, 44, 45]. The leaf water in the vein is less contaminated as well as in stems. The ears of rice and wheat have uptake rates of 30 % (wheat [17, 19]) and 70-100 % (rice [31]). After the exposure during the day end, there is a rapid loss of HTO from leaves with a half-time of 0.5 h in full during the day, but more hours longer in night. For a night exposure, the next morning, there is still 10-20 % HTO in leaves. There is a diurnal pattern of the loss rate. In night conditions, even if the stomata are completely closed, the cuticle conductance varies among plant species and the night uptake is relatively high. The orange and tomato have very low loss rates in night, while rice and wheat have high loss rates. The lettuce data show an interesting pattern; for the young lettuce, the day and night uptake rates are practically the same. For the other crops the night rates are 3-5 times lower than the day ones.

For the formation of OBT, there are problems concerning the extraction of the exchangeable fraction and there is still not a standardized method up to now. Some experimental data show unexpected high non-exchangeable OBT and can be subjected to experimental errors. In the day time, the direct conversion of tritium in OBT through photosynthesis contaminate the assimilate and the further processes of day and night assimilate export to plant parts are less relevant. For night OBT production, the situation is more complex and we find 2 cases (tomato and tangerine) for which the night production is higher than in the day one.

Leaves are the site of OBT production. After one hour exposure, the OBT concentration in water of combustion is up to 1.5 % from the HTO concentration at the end of exposure for the day cases and about 0.1-0.4 % for the night cases (wheat and rice). For plants with growing roots (potato, radish) or reproductive organs (wheat, rice, tomato), the OBT in leaves is exported and there is a loss rate much lower than for HTO. After 6 - 20 hours the OBT concentration in the combustion water in leaf is equal to the HTO concentration and later on is much higher. At harvest, the OBT predominates. For leafy vegetable (and grass perhaps), the OBT in leaves slowly decreases for respiration loss, only. We expect that OBT will predominate after few hours post exposure in the day time and in the next morning for the night exposures. The experimental data are still needed for leafy vegetables and grass.

For longer exposure times, OBT/HTO ratio increases if the environmental conditions are constant.

The experimental data have been obtained in vinyl house or small boxes. Under such conditions the environmental factors affecting tritium transfer and conversion to OBT differs from field.. In the experiments, the forced ventilation was used in order to minimize the boundary and the atmospheric resistance. This increases the both uptake and loss rate. In field condition the exchange velocity and transfer rate between air and plants are lower. In the box experiments, the temperature increases with about 10 °C and in some experiments, the plants have temperature stress and lower uptake and loss rates. As a first approximation we can consider the experimental data valid for field conditions. Due to the diurnal pattern of uptake and conversion to OBT, we expect a diurnal pattern in the ingestion dose and a qualitative results is given in Figure IX.1

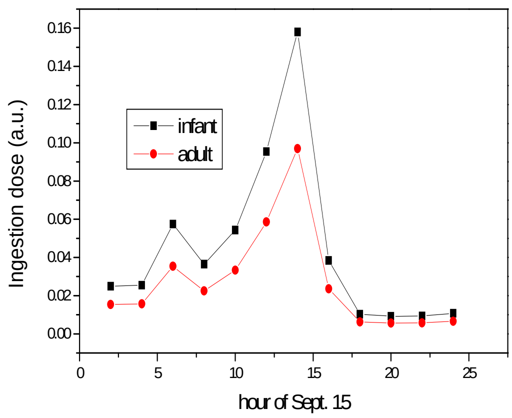


Figure IX.1. Diurnal pattern of ingestion dose (constant air concentration)

The transfer of OBT in edible plant part (translocation index) highly depends on plant development stage and in field conditions the plants have different development stages for the same day of the year. Consequently, we expect a seasonal pattern of ingestion dose and a qualitative picture is given in Figure IX.2.

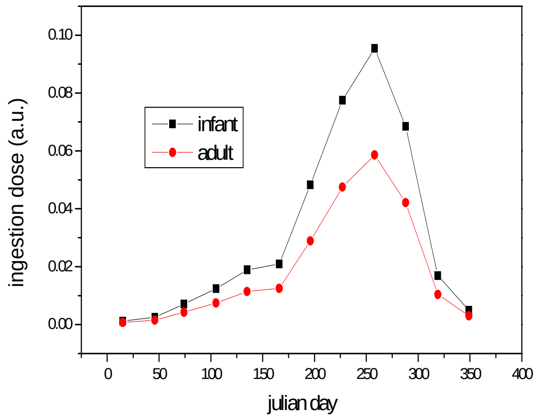


Figure IX.2. Seasonal pattern of ingestion dose (constant air concentration)

The local habits, consumption of local products and climatic conditions influence both diurnal and seasonal pattern (Figures IX.1 and IX.2 are for CANDU reactors in Romania, where the plants and animals are exposed to a constant HTO air concentration for one hour). For CANDU reactors in Romania, the important cultures (decreasing the importance now days) are wheat, maize, sunflower, grapes, vegetable and fruits (apple, peach). Not all of them are found in this overview.

For ITER case, we are interested in hay, corn, wheat, Mediterranean vegetables and crops, grapes, olive, tomatoes, salads, root vegetables (carrots), fruits (apples). In this case we miss the experimental data, also. But it is clear that for ITER, the local consumption of cereals is reduced comparing to the Romanian case and we need more experimental data for grass, salad, olives and grapes. The other needs are for the fuel reprocessing plants in Japan or France.

At the end of this chapter, it is necessary to point one more aspect, related to the natural variability and the experimental data. The measurement of leaves properties demonstrated the large variability of stomata conductance, for the same plant and leave age. When we compared the data with a model results (the best we have) there is a variability of at least a factor 2. In order to define a "reference plant" we must measure a large amount of samples, and this is too costly effective. Generally, about 3 individual plants are measured in a composite sample and this gives a poor image of the real average. Consequently, the experimental results are affected not only by the statistics and sample preparation errors, but also by a systematical error due to few plants analyzed. When we compare the model results with the experimental data, these systematic errors must be considered. In our present understanding, the overall experimental data errors can be at least 50 %, while the analytical and statistical errors are often less than 20 %.

From the modeling view, formation of OBT in day time seems well understood but the processes in night time must be considered in more details in the future. Night OBT cannot be ignored as HTO dispersion in night is weak and concentrations at the receptor can be as high as 40 times day value. Experimental evidences noted above shows that leaf HTO will be only 3-5 times lower that day time with same air concentration. Experimental data shows that TLI in night is close with day values. Consequently, formation of OBT in night can't be excluded in the safety assessment. Our understanding of night process must be enhanced in order to develop robust models. This is a topic to be followed in the next EMRAS.

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