Dynamics of Tritiated Water (HTO) Uptake and Loss by Crops After Short-Term Atmospheric Release

A. J. P. Brudenell,*b C. D. Collins,*b & G. Shaw*

*Centre for Analytical Research in the Environment (CARE), Imperial College at Silwood Park, Ascot SL5 7PY, UK

'Centre for Environmental Technology (ICCET), Imperial College at Silwood Park, Ascot SL5 7PY, UK

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ABSTRACT

A knowledge of the dynamics of HTO in crops and soils is required to validate models, such as the UK Ministry of Agriculture, Fisheries and Food STAR-H3, which predict the ingestion dose following release of tritium. Deposition velocities to cabbage, lettuce and soil have been determined. The loss of HTO from cabbage is described by a double exponential decay; the faster component (ff < 5 min) is attributed to vapour exchange and transpiration, and the slower component (tf > 15 h) to decontamination of stem and root tissues. Due to the long half-time (tf) of the second component, the loss of the last 5% of the applied HTO would appear to be complete within 48 h. This represents a considerable increase over the 6-h loss period predicted by the current STAR-H3 model. As these components can now be resolved from the above data, this may be a useful addition to the STAR-H3 model. Our data indicate that HTO loss predictions should be more pessimistic than the default STAR-H3 parameters allow.

The effect of dark treatment on uptake and loss of HTO has been examined. Pooled data for soils are also well described using a double exponential, fit to the loss data.

Abbreviations:

HTO, tritiated water; OBT, organically bound tritium; HT, tritium gas; OM, organic matter; va, deposition velocity; I, half-time; %RH, percentage relative humidity; STAR-H3, short term atmospheric release of tritium; TFWT, tissue free-water tritium; PAR, photosynthetically active radiation; LAI, leaf area index; MAFF, Ministry of Agriculture, Fisheries and Food; UFOTRI, Unfallfolgenmodell fur Tritiumfreisetzungen (Major German model).
INTRODUCTION

Tritium in the form of tritium gas (HT) and tritiated water (HTO) is released routinely by nuclear installations during normal operation, as well as from accidental releases (Okada & Momoshima, 1993). HTO is the most abundant form of tritium and has been estimated to be 25,000 times more radiotoxic to human beings than HT (ibid.). For this reason, an understanding of the behaviour and fate of HTO in human foodstuffs is of particular importance and forms the basis of this paper.

A knowledge of the dynamics of HTO in crops and soils is required to validate models, such as the UK Ministry of Agriculture, Fisheries and Food (MAFF) STAR-H3 (Smith et al., 1995), which predicts the ingestion dose following the release of tritium to the environment. Current MAFF concerns include the possibility that short discrete discharges have a potential for a significant increase in the public dose. There is a requirement of a better understanding of the dynamics of short-term (24 h or less) tritium discharges to develop a robust assessment methodology.

This study addresses the problem of the dynamics of HTO uptake and loss by crops and soils, and the subsequent fate of tritium in the form of organically bound tritium (OBT) within crop tissues. The subject of tritium in plants, OBT, tritium sampling and measurement, and tritium transport and cycling in the environment have been subject to review (McFarlane et al., 1979; Belot, 1986; Diabate & Strack, 1993; Murphy, 1993; Wood et al., 1993). Studies on temperate food chain contamination by tritium appear to be limited to the grass-milk pathway (Kirchmann et al., 1973) and the vegetable, milk, meat and inhalation routes (Anspaugh et al., 1973). These studies are concerned with doses from chronic exposures to HTO lasting from 7 to 20 days (Kirchmann et al., 1973) and 30 days (Anspaugh et al., 1973), although the latter neglects organically bound tritium. Work on uptake by tomato (Spencer, 1984) maize and barley (Garland & Ameen, 1979; Kim & Baumgartner, 1994)
Tritiated water uptake and loss by crops has been reported. The uptake and loss of HTO by vapour exchange via stomata have been demonstrated for grass and the flux of tritiated water from air to wine grape leaves has been predicted. Studies on the absorption and evaporation of HTO and tritium gas by soil and grassland have been carried out. Assimilation of 3H in leaves of potato and wine grape has also been studied.

Using the above studies as a background to our own work, we have carried out a series of measurements on HTO deposition to and loss from cabbage and lettuce plants and soil surfaces. These measurements have been specifically designed to assess likely tritium ingestion doses following atmospheric releases under UK conditions. Cabbage and lettuce have been chosen as experimental crops because their large edible leaf areas may be expected to maximize the capture of tritiated water vapour. However, the data presented here indicate that this high proportion of leafy tissue would also facilitate subsequent loss of HTO. The tight leaf arrangement in these crops would also limit HTO exchange with internal leaves in the whorl. Lettuce was also selected due to its high water content.

The current STAR-H3 model was designed to minimize the risk of underestimating tritium incorporation into the food chain while avoiding unduly pessimistic assumptions and requirements for detailed input data that may not always be available. Other more complex models that include more detail such as UFOTRI were developed in a research context as well as for regulatory purposes. For example, STAR-H3 does not include atmospheric dispersion, but starts with an assumed tritium concentration in air over agriculturally productive soil. UFOTRI, by contrast, describes the behaviour of tritium in the biosphere and calculates the radiological impact on individuals and the population due to direct exposure and by the ingestion pathways. The compartments of STAR-H3 are illustrated in the conceptual model (Fig. Al, Appendix). The STAR-H3 fast turnover compartment represents the portion of the plant containing water and exchangeable organically bound tritium. The latter is ignored by STAR-H3 as this makes only a minor contribution to the total hydrogen in the compartment. The slow compartment represents the non-labile organically bound tritium (OBT). The tissue free-water tritium (TFWT) loss is modelled by a single exponential function (equation 1)

\[ TFWT_n = a \cdot \exp\left( -\frac{E}{k_n} \right) \]
MATERIALS AND METHODS

Plant material
Cabbage (Brassica oleracea, cv. Greyhound) and lettuce (Lactuca sativa, cv. Diamant) plants were grown from seed in John Innes No. 1 compost. Seedlings were transplanted into John Innes No. 2 compost and grown in an unheated greenhouse under ambient light. Plants were fed every 3 weeks using Phostrogen (NPK 10-10-27; Corwen, Clwyd, UK) as a liquid feed.

Fumigation procedure
Dry air was passed into a Drechsel bottle containing tritiated water (Amersham International plc, UK, 185 MBq ml⁻¹; 0.8 ml made up to 50 ml with tritium-free distilled water) which thus becomes contaminated with HTO vapour. Mature cabbage and lettuce plants were fumigated with this vapour in an enclosed system consisting of a Perspex box inside a controlled environment chamber (Sanyo Gallenkamp, Leicestershire, UK) (Fig. 1). The exhaust air from this was scrubbed using a silica gel trap developed from PVC-U fittings. To maximize air flow, a self-indicating coarse silica gel (BDH/Merck Ltd, Leicestershire, UK) was used in the above trap. The HTO vapour was delivered using a 63-mm-diameter pipe. The Perspex chamber was fitted with inlet and outlet sensors for CO₂, temperature and relative humidity, as well as a PAR sensor, an air temperature thermocouple and three leaf temperature thermocouples. The sensor data were logged on a AT data logger (Delta-T Devices Ltd, Cambridge, UK) and the inlet and outlet CO₂ measured using a WMA-2 analyser and logged using an Egmtrans logger (PP Systems, Hertfordshire, UK). The AT data logger software was set to collect sensor data every 10 s, which were then compressed to give a logged value every 5 min.

Plants were introduced into the Perspex chamber after allowing at least 1 h for the conditions in the controlled environment cabinet to become stable. Plants were then acclimated for at least 12 h. Fumigation was usually carried out for 1 h.

The organic matter of selected soils was determined by weight loss following combustion; soil samples were passed through a 3-mm sieve, oven-dried overnight and combusted at 400°C (1 °C min⁻¹ ramp; Carbolite OAF 1000, Fisons, UK) for 4 h. The soils selected for fumigation were 'Silwood Orchard' [mesozoic and tertiary sand and loam; 7.43% organic matter (OM)] from Silwood Park ground, sand and two soils (Sl: glacio-fluvial drift; 17.6% OM; S2: chalky till and glacio-fluvial drift; 5.74% OM)
Tritiated water uptake and loss by crops

Fig. 1. Schematic of fumigation set up, which consists of a Perspex chamber inside a controlled environment cabinet. To ensure mixing of inlet gases, two electric fans are attached to a grille just after the point of entry of the inlet pipe into Perspex chamber. The maximum air velocity in chamber was 0.44 m s⁻¹ and the velocity at canopy height was 0.24 m s⁻¹. The position of the environmental sensors and the data flow to the logger are indicated.

Soil samples for fumigation were prepared by passing through a 2-mm sieve into plastic trays to a depth of 1 cm. These were wetted to field capacity then allowed to drain prior to a deposition experiment. After each fumigation, samples were taken as 6.5-cm-diameter zones, which were then weighed and subjected to the extractions for tritium analysis below.

Dark treatment
Lettuce plants were subjected to dark treatment before and during HTO fumigation. HTO fumigation was carried out 2 h after placing plants in darkness. Control plants were fumigated in light and moved into ambient air outside in light. Dark treated plants were either left in the dark in the Perspex chamber with clean air flow to purge the fumigation system of HTO, or were transferred into the light as for controls. To check the effect of dark on loss following uptake in the light, cabbage plants were fumigated for 1 h in the light followed by 1 h fumigation in the dark before sampling. This was to prevent loss occurring before stomata were closed. Unlike the lettuce experiment above, these cabbage plants were subjected to dark treatment during HTO fumigation at dusk to avoid upsetting the plants' diurnal rhythm.
Stomata1 resistance

Values for stomata1 resistance of cabbage plants under greenhouse condition, in controlled monitored conditions in the fumigation chamber in both light and dark were determined using a AT AP4 Porometer (Delta-T Devices Ltd). Standard calibration procedures were followed each time a set of measurements was made.

HTO recovery

Sampling for HTO recovery was performed on duplicate plants. Most experiments were repeated three times with similar results. Fumigated plants were weighed and put either into plastic bottles or into plastic bags for storage at -20°C. Fifty millilitres of tritium free distilled water was then added to those plants put into plastic bottles. These were then placed on to a flask shaker for 24 h to allow extraction of HTO (Amano & Garter-r, 1991). Soils were treated in a similar manner, following the method of Garland (1980), except that samples were extracted overnight (16 h).

Before determining non-labile OBT, tissue water and HTO were also removed by freeze-drying and trapping using liquid nitrogen. The tissue water and HTO was collected in a specially made double arm glass cold trap (AS0 Glassblowing Ltd, Oxfordshire, UK). The freeze-drying apparatus was developed in collaboration with RCD Ltd (Radiocarbon Dating Ltd, Oxfordshire, UK) and was based on their design. A direct double vacuum pump (Model DDL40; Javac (UK) Ltd, Middlesbrough, Cleve-

Tissue oxidation

Once all the HTO had been removed from samples by freeze-drying, the exchangeable OBT was removed by oven drying, equilibrating with moist air and subsequent oven re-drying. The remaining non-exchangeable OBT was then determined using tissue oxidation in a stream of oxygen in an automatic sample oxidiser (Model 307; Canberra-Packard, Pangbourne, Berkshire, UK).

Scintillation counting

Tritium levels were determined by scintillation counting for 1Omin using Optiphase 'HiSafe' 3 (Wallac, Milton Keynes, UK) scintillation cocktail for HTO samples and Monophase S (Canberra-Packard) for oxidized samples. Scintillation counting was performed using a Wallac 'Rackbeta'
Deposition velocity

The deposition velocity ($v$) for HTO was calculated, after Garland (1980), using eqn (2).

$$HTO \text{ deposited (Bq m}^{-2}) = \text{concentration in air (Bq m}^{-3}) \times \text{exposure time (s)}$$

Data analysis and curve fitting

The data analysis and curve fitting were carried out using a graphics and statistics package 'Prism' (Version 2.0, GraphPad Software, San Diego, CA, USA); the iterative nonlinear regression module allowing the determination of the comparative goodness of fit of two equations to the same data set. In testing for a better fit, the more complex double exponential was chosen when $P < 0.05$ when compared with a single exponential and where the decrease in the sum of squares is worth the 'cost' of the additional variables (loss of degrees of freedom). The program was also used to check for deviation from the chosen equation by means of a Runs test, and all the fitted curves reported here did not systematically differ from the data.

RESULTS AND DISCUSSION

HTO recovery methods

The recovery of tissue water and the potential for isotope effects during freeze-drying, vacuum distillation and azeotropic distillation with regard to enrichment of OBT have been extensively examined by Kim and Baumgartner (1990, 1991, 1994) and Kim et al. (1991). From their work, it could be expected that greater recovery would be found for HTO with extraction using distilled water. A bias in favour of HTO recovery by extraction with distilled water has been demonstrated with maple and hickory foliage (Amano & Garten, 1991). A linear relationship with a slope of 0.8 has been determined for HTO recovery from extraction with distilled water vs. HTO recovery by freeze-drying (Fig. 2). Further work will be required to confirm Amano's data.
Air concentration time course

Time course studies of HTO vapour concentrations from the chamber air sample line indicate that the chamber environment is consistent during fumigation, with a steady state concentration being established after \( \tau = 10 \) min fumigation. The presence of plants has a small effect on this concentration (Fig. 3).

Partitioning between tissues

Partitioning of HTO between different tissues has been examined (Fig. 4). Division of cabbage plants into stem and leaf whorls indicates a lower rate of HTO uptake by stems, with less tritium partitioning to the inner leaves. The fresh weights of the above plants averaged \( 34.89 \pm 4.32 \) g.
Tritiated water uptake and loss by crops

Fig. 3. Time course of air sample line during HTO fumigation with and without lettuce plants. The 30-min fumigations were performed using the same source of HTO, while the 120-min fumigation was performed using a relatively depleted source as this fumigation was intended solely as a check on chamber behaviour.

Deposition velocities

HTO deposition velocities (vs) for lettuce, cabbage and soil (Silwood Orchard) are given in Table 1. These compare reasonably with the range 4 x 10^{-4} to 4 x 10^{-3} ms^{-1} for soil and typical daytime values for vegetation of 2.5 x 10^{-4} ms^{-1} (Bunnenberg et al., 1990) and the range 1.2 x 10^{-4} to 1.4 x 10^{-4} ms^{-1} determined for vegetation by Mason et al. (1973).

Loss rates from plants

Loss rates have been determined for plants fumigated in the Perspex chamber and then moved outside into ambient air (Fig. 5) for which...
Fig. 4. Partitioning of HTO to different cabbage tissues following a 30-min fumigation. The outer most whorl is no. 1 and the inner most is no. 4.

TABLE 1

| Material  | Deposition Velocity (vg) |
|-----------|--------------------------|
| Cabbage   | 3.99 x 10^{-3} 4.82 x 10^{-4} |
| Lettuce   | 1.25 x 10^{-3} 1.63 x 10^{-4} |
| Soil      | 4.94 x 10^{-4} 7.16 x 10^{-5} |

Meteorological conditions are listed in Table A1 (Appendix) for the period of HTO loss. Control plants not exposed to HTO were found to have tritium levels indistinguishable from background. As described in the figure legend, the loss of HTO was found to be due to two components. The faster component \( t' \sim 53 \text{ min} \) is attributed to loss
Tritiated water uptake and loss by crops

I I I 1
TFTW, = a.eqYk+'+b.expsk2.'+ p
tx (k,)=0.88h (53 min)
tK (k,)=152h
bar+SEM

0 6 12 18 24 30 36

Time (hours) after end of fumigation

Fig. 5. Loss of HTO from cabbage in ambient air following a 60-min fumigation. The data are described by a double exponential decay, from which rate constants \( k_t \) and \( k_2 \) are derived. The half-time \( t_{50} \) for loss due to \( k_1 \) is estimated as 53 min and the half-time \( t_{50} \) of loss due to \( k_2 \) is estimated as 152 h. The inset graph further confirms the existence of these two linear components, the lines fitted by least squares regression. These are taken to represent the loss due to (i) vapour exchange and transpiration and (ii) decontamination of stem and root tissue. Meteorological data for the HTO loss period are listed in Table A1 (Appendix).

due to vapour exchange and transpiration (discussed later). Belot et al. (1979) concluded that evidence of vapour exchange of leaf water with atmospheric water has been substantiated by several observations (Vaadia & Vaisel, 1963; Kline & Stewart, 1974). The slower component \( t_{50} 15 \) h) is attributed to decontamination of stem and root tissues, as postulated by Guenot and Belot (1984) that have been labelled by phloem translocation (Biddulph & Cory, 1957; McFarlane et al. 1979; Couchat et al., 1983). This is a less significant component in terms of the overall amount of HTO lost, but contributes significantly to the extension of the period over which total loss process occurs. With regard to the identification of this stem/root decontamination mechanism, it is of interest that, following a 1-h HTO exposure to Medicago hispida (Burclover), Koranda and Martin (1973) identified a further loss phase with a much longer \( t_{50} \) of 270 h, attributed to organically fixed tritium.

The slower stem/root decontamination component should not be confused with the 'slow turnover' compartment of STAR-H3 which is...
concerned with non-labile OBT formation. Thus the 'fast turnover' compartment of STAR-H3 could be expanded to include the slower stem/root decontamination process or further compartments added to the model.

Preliminary results from short term HTO loss from passing air at 100% RH or 0% RH over a single contaminated cabbage plants (and bare soil) have allowed the resolution of the above loss mechanisms of vapour exchange ($t; M \approx 2\text{min}$) and transpiration ($I \approx 60\text{min}$) (Brudenell, unpublished). The process of vapour exchange would be quickly (<20min) subsumed by transpiration losses, and hence vapour exchange is not resolvable in the medium term experiments presented here. These data largely confirm the conclusion that, at least initially, the HTO exchange mechanism is dominant over transpiration (Smith et al. 1995). In the above fumigation, 95% of the HTO was lost in 6 h (i.e. seven half-times of component 1). In two half-times of component 2 (30 h), the tissue free-water tritium (TFWT) was reduced to 1%. These results extend the work of Belot et al. (1979) who obtained similar half-times for the first component, but were unable to resolve the second component because of the short duration of their experiments.

The fast compartment water turnover rate in the STAR-H3 model is set at 1 h$^{-1}$ (Smith et al., 1995). Due to the long half-time of the second component, the loss of the remaining 5% labile HTO would appear to be complete within 48 h. This represents a considerable increase over the 6 h loss period predicted by the current STAR-H3 model. As these components can now be resolved from the above data, this may be a useful addition to the STAR-H3 model.

Anspaugh and co-workers (1973) obtained a $t_f$ of 45 min for the first component and a $t_I$ of 25 h for the second component for native species Medicago hispida and Amsinckia grandis following a 30-min exposure to HTO. However, there was a difference of nearly an order of magnitude between the two species for the tritium concentration for the second component (ibid.) Koranda and Martin (1973) obtained half-times of 20 and 55 min for the first component for sunflower and red oat (Avena) respectively after a 5-h fumigation. Using a 4-h HTO exposure period, Guenot and Belot (1984) obtained a $t_I$ of 30 min for the loss of HTO from tissue water of potato and grape leaves and a $t_I$ of 30 h for a second component. It should be noted that it is unclear as to how the above literature values for half-times of loss were derived, and the equations fitted to the data are not given. From the data in this study, it can be seen that the half-times for cabbage are slightly longer for the first component, with a correspondingly shorter half-time for the second component (Fig. 5).
Rates of loss from soils via volatilization from soils, have been determined for chamber fumigated soils moved outside into ambient air (Fig. 6) for which meteorological conditions are listed in Table A2 (Appendix) for 0.0 1 1 1 360 720 1080 1440 0 360 720 1080 1440 Time (min) after end of fumigation.

Fig. 6. Loss of HTO from soils into ambient air, following fumigation in the chamber. The data for the individual soils (b) can be described with a single exponential decay curve, giving a range of similar half-times of loss. The upper graph (a) is for the pooled data for all soil types. The solid line is a double exponential curve similar to that in Fig. 5. The half-time for rate $k_1$ is 25 min, and that for $k_2$ is 28.4 h. S1,2 = soil samples taken from Thompson and Morgan Ltd as described in the Materials and Methods. Meteorological data for the HTO loss period are listed in Table A2 (Appendix).
A. J. P. Brudenell et al.

The period of HTO loss. As described in the figure legend, data for individual soils can be described using a single exponential decay. However, the pooled data for all soil types are well described using a double exponential curve. The fast loss rate would appear to be a diffusion process not necessarily linked to water loss (Garland & Cox, 1980) especially immediately after the end of a fumigation. However, vapour exchange is assumed to be confounded with evaporation as discussed above. The rates for the first component are shorter for cabbage (Fig. 5) although the half-time for the second component was found to be slightly longer. This is in agreement with Garland and Cox (1980) who found that the presence of grass appeared to introduce an additional resistance and reduce the rate of exchange at the surface. No relationship was discernible between the amount of HTO taken up by each soil type and the organic matter content, one justification for pooling soil data. This factor and the longer half-time for the second component may reflect a tendency for a fraction of the applied HTO to become associated with water more closely bound to soil particles or other less accessible zones of water in the soil.

Environmental conditions

That the chamber environmental conditions are well controlled is demonstrated by the logger data (Fig. 7). The perturbations are due to sampling events within the chamber. The fast recovery following sampling and maintenance of chamber settings over >24h is also demonstrated. The cabbage data included with logger data are for HTO loss in the light under the monitored conditions.

The meteorological data (Appendix Tables A1 and A2) for the HTO loss period of both cabbage (Fig. 5) and soil (Fig. 6) indicate that, for the first 6 h, when the most rapid loss occurred, environmental conditions were fairly stable. This is most noted for wind speed, air temperature and %RH. This was taken to be advantageous as these factors could markedly affect the rate of loss of HTO.

Effect of dark treatment on HTO loss

The effect of dark treatment following uptake in the light can be compared with loss of HTO in the light (Fig. 8). The light 'control' data are given in Fig. 7. Cabbage fumigated under the same conditions was allowed to lose HTO in the dark. The stomata conductance in the light was 627.5 cm\(^{-2}\) sl-1, slightly higher than the UFOTRI default of 400 cm\(^{-2}\) sl-1, whilst the LAI for cabbage was 2.94, similar to the UFOTRI default of
Tritiated water uptake and loss by crops

Time (min)

Fig. 7. Logger data of environmental conditions in Perspex chamber during loss of HTO from fumigated cabbage. The leaf area index (LAI) of plants in the chamber was approximately 2.94.

0 6 12 18 24 30 36 42

Time (hours) after end of fumigation

Fig. 8. Effect of dark on loss of HTO from cabbage. The 'control' data set for light conditions is as given in Fig. 7 which also includes the logged environmental data from the sensors in the Perspex chamber. Only one half-time ($t_1$) is resolvable in the dark, while in light, two half-times ($t_2$, $t_3$) are resolvable. The stomatal resistance of plants under greenhouse conditions averaged 670 ± 40 s·m⁻¹, under monitored chamber conditions 627.5 ± 199.7 s·m⁻¹ and after 1 h of darkness 3120 ± 835 s·m⁻¹ (Raskob, 1990). Two loss rates were resolvable in the light, while only one loss rate was resolvable for loss in the dark. The rate of loss in the dark is reduced ($t_2 = 13$ h) compared to the faster half-times for loss in light ($t_2 = 5.4$ mm, $t_3 = 6.7$ h). This is in reasonable agreement with
the data of Kline and Stewart (1974) regarding grass, obtaining a t; of 35 f 5 min in the light and the dark.

The effect of dark treatment on both uptake and loss of HTO has been examined (Fig. 9). Uptake via stomata is obviously important, as indicated by the low contamination levels for cabbage fumigated in the dark (inset graph). The apparent lack of any effect of light treatment following the dark treatment (compare D, with L,) could indicate that stomata were not fully closed as, unlike plants fumigated in the dark (inset graph), these lettuce plants were placed in darkness during daylight.

Fig. 9. Effect of dark treatment on uptake of HTO by lettuce. Dark-treated plants were fumigated with HTO, after which plants were either left in the dark (D, -s) or moved into the light (L, -z). The controls were treated in the light as for routine fumigations. The dark treatment reduced the uptake of HTO (Or). Loss of HTO did not appear to be affected by light treatment after dark treatment (compare D, with L,). After 1440 min (24 h), HTO levels were the same in the control and treated plants. The inset graph shows the data from lettuce fumigated in the dark and left to lose HTO in the dark.
Tritiated water uptake and loss by crops 213 hours (no porometer readings available for these data). This would appear to be more likely as substantial labile surface contamination was not observed for plants fumigated in the dark (inset graph). This result may be a useful warning against attempting to work against the plants’ diurnal rhythm.

Non-exchangeable OBT

The build up of non-exchangeable OBT can be seen to increase over time following a 1-h fumigation (Fig. 10). This would indicate the formation of OBT metabolically (including isotopic exchange with relocation to a non-exchangeable hydrogen position by enzymic reactions (Diabate & Strack, 1993)) rather than enhanced OBT due to artifactual tritium enrichment of tissue water during freeze-drying, as no increase over time would be expected with the latter. The partitioning of OBT (Fig. 10a) would appear to follow the pattern of HTO levels shown in Fig. 4. The formation of non-labile OBT would appear to be slow as indicated by the low values up a)

Fig. 10. Levels of fixed-OBT determined by tissue oxidation of freeze-dried cabbage plants after HTO fumigation: (a) 1 day after fumigation and (b) at intervals up to 30min, 1 day and 3 days after fumigation.
A. J. P. Brudenell et al. to 30 min, although following an increasing trend. The lower levels after 3 days may indicate turnover, although further work will have to be done to confirm this observation. The low level detected in the roots may be due to the low exposure of this tissue or may indicate effective decontamination (Fig. lob).

CONCLUSIONS

The results from this study demonstrate that crop plants, such as cabbage and lettuce, once exposed to HTO vapour, effectively decontaminate themselves in the light and dark, although at a reduced rate in the latter. The applied HTO is almost completely absent 48 h after a short term fumigation. The tissue free-water tritium (TFWT) loss is well described by a double exponential function (eqn (3)) rather than the current STAR-H3 single exponential function (eqn (1)).

$$\text{TFWT,} = a \cdot \exp(K' \cdot t) + b \cdot \exp(-K2 \cdot t)$$

Our data indicate that HTO loss predictions should be more pessimistic than the default STAR-H3 parameters allow. The fit of a double exponential relationship with similar rate constants to the data from both soils and crops would indicate that the processes involved are mainly physical. The fit to our data of the more complex double exponential was chosen when $P < 0.05$ when compared with a single exponential. The fitted curves reported here did not differ systematically from the data when checked with a Runs test. The cited literature is often unclear regarding the fitted equation and method used to derive a half-time for a loss process. However, it would be expected that species/crop plant differences, such as stomata conductance or the use of different experimental setups and conditions, are the major cause of the range of loss rates seen in the literature. The plants' biochemistry is of significance when considering the fate of tritium as fixed-OBT. This is of interest with regard to the dose estimates arrived at through the use of models such as STAR-H3 or UFOTRI. Further work on the dynamics of HTO in crops (including apple and potato), and an appraisal of the experimental methodology and HTO application methods are planned.

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### APPENDIX

**TABLE A1**

Meteorological Data for HTO Loss from Cabbage (Loss Data in Fig. 5)

| Time (h) | Air temp. (°C) | Relative humidity (% RH) | Solar radiation (W m⁻²) | Wind speed (m s⁻¹) |
|----------|----------------|--------------------------|--------------------------|------------------|
|          |                |                          | Min | Max | Mean |                |                  |
| 13-00    | 28.54          | 0.21                     | 39.00 | 1.00 | 823.00 | 861.00 | 841.80 | 1.71 | 0.73 |
| 14-00    | 28.16          | 0.43                     | 38.00 | 1.00 | 747.00 | 807.00 | 778.30 | 1.71 | 0.48 |
| 15-00    | 28.43          | 0.19                     | 36.00 | 0.00 | 629.10 | 719.00 | 675.00 | 1.22 | 0.17 |
| 16-00    | 28.12          | 0.51                     | 35.00 | 1.00 | 488.90 | 593.60 | 540.70 | 1.12 | 0.28 |
| 17-00    | 26.33          | 1.45                     | 40.00 | 3.50 | 115.40 | 449.50 | 302.30 | 0.93 | 0.18 |
| 18-00    | 23.25          | 0.73                     | 50.00 | 4.50 | 113.90 | 248.00 | 180.10 | 0.90 | 0.36 |
| 19-00    | 20.45          | 1.14                     | 62.00 | 3.50 | 0.00   | 67.80  | 31.28  | 0.73 | 0.21 |
| 20-00    | 17.32          | 0.99                     | 73.00 | 3.50 | 0.00   | 0.00   | 0.00   | 0.61 | 0.20 |
| 21-00    | 14.76          | 0.89                     | 81.00 | 3.00 | 0.00   | 0.00   | 0.00   | 0.39 | 0.11 |
| 22-00    | 12.89          | 0.47                     | 87.00 | 2.00 | 0.00   | 0.00   | 0.00   | 0.33 | 0.08 |
| 23-00    | 11.89          | 0.47                     | 91.00 | 2.00 | 0.00   | 0.00   | 0.00   | 0.34 | 0.10 |
| 24-00    | 10.75          | 0.33                     | 96.00 | 1.50 | 0.00   | 0.00   | 0.00   | 0.28 | 0.12 |
| 1-00     | 10.21          | 0.24                     | 98.00 | 1.00 | 0.00   | 0.00   | 0.00   | 0.27 | 0.17 |
| 2-00     | 9.72           | 0.17                     | 99.00 | 0.00 | 1.00   | 14.05  | 7.25   | 0.13 | 0.03 |
| 3-00     | 9.94           | 0.52                     | 99.00 | 0.00 | 20.50  | 50.50  | 32.70  | 0.08 | 0.09 |
| 4-00     | 11.38          | 0.39                     | 99.00 | 0.00 | 69.20  | 90.60  | 81.78  | 0.35 | 0.07 |
| 5-00     | 12.32          | 0.40                     | 99.00 | 1.50 | 105.50 | 201.30 | 142.00 | 0.54 | 0.77 |
| 6-00     | 13.31          | 0.25                     | 93.00 | 1.50 | 299.30 | 588.60 | 484.90 | 0.60 | 0.15 |
| 7-00     | 15.10          | 1.35                     | 84.00 | 6.00 | 589.20 | 684.20 | 638.20 | 0.93 | 0.18 |
| 8-00     | 20.01          | 1.02                     | 66.00 | 3.50 | 705.00 | 778.00 | 740.80 | 0.92 | 0.36 |
| 9-00     | 22.27          | 1.14                     | 59.00 | 3.00 | 801.00 | 847.00 | 824.30 | 1.20 | 0.29 |
| 10-00    | 22.15          | 1.01                     | 51.00 | 3.00 | 851.00 | 859.00 | 853.80 | 1.18 | 0.23 |
| 11-00    | 27.38          | 1.02                     | 51.00 | 3.00 | 815.00 | 837.00 | 826.00 | 1.27 | 0.16 |
| 12-00    | 29.26          | 0.41                     | 46.00 | 2.00 | 740.00 | 827.00 | 772.80 | 1.24 | 0.20 |
| 13-00    | 30.35          | 0.71                     | 43.00 | 0.50 | 662.70 | 680.10 | 670.40 | 1.22 | 0.21 |
| 14-00    | 30.40          | 0.48                     | 41.00 | 0.00 | 526.40 | 616.80 | 571.40 | 2.01 | 0.16 |
| 15-00    | 30.05          | 0.21                     | 43.00 | 0.00 | 397.10 | 508.60 | 452.20 | 1.65 | 0.44 |
| 16-00    | 29.39          | 0.38                     | 42.00 | 0.00 | 119.50 | 235.10 | 176.00 | 1.92 | 0.13 |
| 17-00    | 27.69          | 0.51                     | 40.00 | 0.00 | 57.90  | 216.90 | 119.30 | 1.83 | 0.35 |
| 18-00    | 25.96          | 0.37                     | 36.00 | 1.00 | 12.40  | 67.80  | 32.38  | 1.85 | 0.61 |
| 19-00    | 24.61          | 0.60                     | 36.00 | 0.50 | 0.00   | 5.90   | 1.65   | 1.07 | 0.34 |
| 20-00    | 22.35          | 1.13                     | 37.00 | 0.00 | 0.00   | 0.00   | 0.00   | 0.42 | 0.29 |
| 21-00    | 18.66          | 1.58                     | 40.00 | 2.00 | 0.00   | 0.00   | 0.00   | 0.05 | 0.04 |

Values are averages for measurements logged at 15-min intervals.
TABLE A2
Meteorological Data for HTO Loss from Soil (Loss Data in Fig. 6)

| Time (h) | Air temp. (°C) | ± Relative humidity (%RH) | Net radiation (W m²) | ± Wind speed (m s⁻¹) |
|---------|----------------|---------------------------|----------------------|---------------------|
|         |                |                           | Min                  | Max                 | Mean                |
| 13:00   | 19.00          | 2.50                      | 58.00                | 7.00                | 82.00               | 60.00               | 250.00              | 0.67               | 0.19               |
| 14:00   | 18.00          | 0.50                      | 58.00                | 3.50                | 140.00              | 280.00              | 200.00              | 0.65               | 0.23               |
| 15:00   | 19.00          | 0.00                      | 54.00                | 1.50                | 180.00              | 210.00              | 190.00              | 0.78               | 0.19               |
| 16:00   | 19.00          | 0.50                      | 49.00                | 3.00                | 84.00               | 220.00              | 170.00              | 0.66               | 0.14               |
| 17:00   | 18.00          | 0.00                      | 53.00                | 1.00                | 69.00               | 89.00               | 80.00               | 0.62               | 0.13               |
| 18:00   | 17.00          | 0.50                      | 54.00                | 2.00                | 58.00               | 140.00              | 90.00               | 0.63               | 0.06               |
| 19:00   | 17.00          | 0.50                      | 54.00                | 3.00                | 35.00               | 81.00               | 56.00               | 0.41               | 0.19               |
| 20:00   | 16.00          | 1.00                      | 61.00                | 4.50                | -21.00              | 30.00               | 8.60                | 0.56               | 0.19               |
| 21:00   | 14.00          | 0.50                      | 73.00                | 3.00                | -36.00              | -18.00              | -24.00              | 0.78               | 0.23               |
| 22:00   | 13.00          | 0.50                      | 76.00                | 1.50                | -24.00              | -21.00              | -23.00              | 0.34               | 0.25               |
| 23:00   | 12.00          | 0.00                      | 79.00                | 4.00                | -23.00              | -21.00              | -22.00              | 0.25               | 0.18               |
| 24:00   | 9.70           | 1.80                      | 89.00                | 5.50                | -56.00              | -25.00              | -47.00              | 0.14               | 0.13               |
| 1:00    | 5.80           | 0.90                      | 98.00                | 1.00                | -45.00              | -35.00              | -39.00              | 0.00               | 0.00               |
| 2:00    | 4.50           | 0.15                      | 99.00                | 0.00                | -34.00              | -32.00              | -33.00              | 0.00               | 0.00               |
| 3:00    | 4.10           | 0.15                      | 99.00                | 0.00                | -33.00              | -27.00              | -31.00              | 0.00               | 0.00               |
| 4:00    | 5.00           | 0.85                      | 99.00                | 0.00                | -24.00              | -17.00              | -20.00              | 0.03               | 0.03               |
| 5:00    | 5.30           | 0.20                      | 99.00                | 0.00                | -24.00              | -19.00              | -22.00              | 0.02               | 0.02               |
| 6:00    | 5.70           | 0.85                      | 99.00                | 0.00                | -16.00              | 22.00               | 4.80                | 0.02               | 0.02               |
| 7:00    | 8.60           | 0.60                      | 98.00                | 1.00                | 25.00               | 49.00               | 34.00               | 0.14               | 0.14               |
| 8:00    | 11.00          | 1.15                      | 91.00                | 8.00                | 59.00               | 94.00               | 74.00               | 0.04               | 0.04               |
| 9:00    | 13.00          | 0.50                      | 78.00                | 3.00                | 98.00               | 180.00              | 120.00              | 0.21               | 0.21               |
| 10:00   | 15.00          | 1.50                      | 65.00                | 10.50               | 150.00              | 310.00              | 220.00              | 0.12               | 0.12               |
| 11:00   | 15.00          | 1.00                      | 62.00                | 4.50                | 57.00               | 180.00              | 110.00              | 0.11               | 0.11               |
| 12:00   | 16.00          | 0.50                      | 56.00                | 1.00                | 130.00              | 220.00              | 180.00              | 0.11               | 0.10               |

Values are averages for measurements logged at 15-min intervals.

Fig. A1. Conceptual model for incorporation of tritium into the terrestrial food chain (adapted from Smith et al. (1995)). Compartments of relevance to this report are emphasized. The conceptual model was based on a review of existing models, relevant available data, and MAFF's particular assessment requirements (ibid.). For reasons of clarity, reference to tritium gas (HT) in the original diagram has been omitted, but this does not imply any lack of consideration of HT behaviour in the STAR-H3 code.