Deposition of gaseous radionuclides to fruit

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Abstract

$^{14}$C, $^{35}$S and $^{3}$H are released to the environment during the operation of gas-cooled reactors and were identified as radionuclides of interest by the BIOMASS Fruits Working Group. This paper provides a review of the deposition, uptake, allocation and loss of these radionuclides with respect to fruit and conceptual models for gaseous radionuclides. It is concluded that the mechanisms for the uptake of $^{35}$S, HTO and $^{14}$CO$_2$ are well understood and that their deposition velocities have been quantified. There is also a reasonable body of work on the translocation of $^{14}$C once in the crop, but much less for $^{35}$S and $^{3}$H, which are considered to follow source-sink relationships. The loss rates of the three radionuclides show large differences, with tritium lost rapidly in the form of HTO but retained longer when converted to OBT. The losses of $^{14}$C are less and those of sulphur are minimal post fixation. When fruit crops alone are considered, the quantity of information is further reduced but predictions on possible behaviour of these radionuclide species can be made from the current knowledge. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

During the routine operation of nuclear installations such as UK Magnox and advanced gas cooled (AGR) reactors, and pressurised water reactors (PWR), which are more common in the rest of the world, the radionuclides $^{14}$C, $^{35}$S and $^{3}$H are continually released to the environment (Brudenell et al., 1997; Collins, 1994; Collins & Gravett, 1995; Okada & Momoshima, 1993). The principal species are of

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[14C]-carbon dioxide, [14C]-methane, [35S]-hydrogen sulphide, [35S]-carbonyl sulphide, [3H]-water (HTO), [3H]-hydrogen (HT) and [3H]-methane. In addition to these chronic discharges, spike discharges occur during certain reactor operations known as ‘blowdown’, where the CO2-coolant gas in the reactor circuit is depressurised and released to the environment prior to shutdown of the reactor, and ‘start up’, when the reactor goes critical following a shutdown (Collins & Gravett, 1995). The present review aims to determine the key processes for the behaviour of these radionuclides in fruit. The review is divided into three sections; uptake, allocation and loss, with the aim of providing information on key processes and, where possible, data to allow for the development of a conceptual model.

2. Uptake

Plant canopies constitute a major sink for radioactive gaseous pollutants, and uptake of these by vegetation may play an important part in the subsequent ingestion dose to man following such releases. Whilst the physical and chemical processes controlling such deposition are well characterised, the processes operating at the leaf surface and within the leaf interior are not always so well understood. Additionally, although uptake by vegetable crops has been studied fairly extensively, there has been relatively little investigation into uptake by fruit crops. The following paragraphs review the uptake of 14C, 35S and 3H by crops. Where possible, reference is made to fruit crops and fruit trees, otherwise the behaviour in other plants and ecosystems is used to illustrate principles that may be applied to fruit.

2.1. Deposition

Foliar uptake is the principal pathway for the uptake of radioactive gases by vegetation, usually through small pores in the leaf surface known as stomata. The process of gas exchange between plants and the atmosphere can be broken down into three components (Kurzeja, Murphy & Taylor, 1988; Fowler, 1987): i.e. transfer from a reference height through the free atmosphere to within a few millimetres of the surface, transfer through the viscous sublayer near the leaf surface, and finally, surface capture of the gas. The exchange process is often modelled with a ‘resistance’ analogue method, which allocates an ‘exchange resistance’ to each of the above parts (Wesely and Hicks, 1977). Deposition of gases to plants is often described by a resistance model where the aerodynamic (r_at), the boundary layer (r_bu) and the stomatal resistances (r_st) determine the fluxes at the leaf’s surface layer (Fig. 1). The sum of the three resistances (units: s m−1) yields the total resistance whose reciprocal is the deposition velocity of the gas (Table 1). Thus, the deposition rate equals the surface concentration divided by the total resistance of the pathway to the vegetation from the atmosphere (Kurzeja et al., 1988). Kurzeja et al. (1988) and Murphy and Corey (1975) found that the controlling step for both HTO uptake from and loss to the atmosphere for plants appears to be flux through stomata. The stomatal resistance is also known to dominate for CO2 and COS (Unsworth, 1981; Collins & Scarisbrick, 1994). For instance,
Fig. 1. Resistance analogue, showing resistance towards the leaf surface showing boundary layer ($r_{bv}$), stomatal ($r_{st}$), cuticular ($r_{c}$), intercellular ($r_{i}$), wall ($r_{w}$) and leaf ($r_{l}$) resistances.

Table 1
Deposition Velocity of CO$_2$, HTO and COS to vegetation, showing measurements from field (F), laboratory (L) and wind tunnel (T) and dark (D)

| Gas     | Deposition velocity (cm s$^{-1}$); Location; (crop) | Reference                           |
|---------|-----------------------------------------------------|-------------------------------------|
| CO$_2$  | $2 \times 10^{-3}$–$8 \times 10^{-3}$ F ($Fagus sylvatica$)  |
|         | $6 \times 10^{-3}$–$1.2 \times 10^{-3}$ F ($Glycine max$)  |
|         | $1.4 \times 10^{-2}$–$2.8 \times 10^{-2}$ F ($Zea mays$)  |
|         | $4.3 \times 10^{-2}$ F ($Phaseolus vulgaris$)            |
|         | $3.6 \times 10^{-2}$ ($Glycine max$)                     |
|         | $7 \times 10^{-2}$ (Spring wheat)                       |
|         | 2.5 – $10^{-2}$ (range $10^{-3}$–$5 \times 10^{-2}$)    |
|         | $1.2 \times 10^{-2}$–$1.4 \times 10^{-2}$               |
|         | $3 \times 10^{-2}$–$6 \times 10^{-3}$                   |
|         | $4.0 \times 10^{-7}$ F (soybean)                        |
|         | $6.4 \times 10^{-8}$ F (corn)                           |
|         | $2.3 \times 10^{-8}$ F (white spruce)                   |
|         | $7.3 \times 10^{-8}$ F (red maple)                      |
| HTO     | $8.4 \times 10^{-2}$ L ($Lolium esculentum$)             |
|         | $3.1 \times 10^{-2}$ D ($Lolium perenne$)                |
|         | $8.3 \times 10^{-3}$ F ($Lolium perenne$)                |
|         | $4.6 \times 10^{-3}$ FD ($Lolium perenne$)               |
|         | $5.2 \times 10^{-2}$ L (cabbage)                         |
|         | $5.7 \times 10^{-2}$ L (radish)                          |
|         | $5.6 \times 10^{-2}$ L (turnip)                          |
|         | $8.0 \times 10^{-2}$ T ($Lolium perenne$)                |
|         | $3.1 \times 10^{-2}$ TD ($Lolium perenne$)               |

Salisbury and Ross (1992) pers. comm.
Winzeler (1979)
Bunnenberg, Taschner and Ogram (1990)
Mason et al. (1973)
Higgins, Shaw, Haywood and Jones (1996)
Spencer, Ogram and Brown (1988)
Taylor, McLaughlin, shriner and Selvidge (1983)
Kluczewski, Brown, Bell and Minski (1985)
Brown, Kluczewski and Bell (1986)
Brown et al. (1986)
Chadwick (1977)
Biscoe, Scott and Monteith (1975) recorded no net uptake of $^{14}$CO$_2$ in wheat in the dark, when stomata are usually closed for the majority of British crops (Salisbury & Ross, 1992). Thus, environmental conditions resulting in a low stomatal resistance, i.e. high light and plentiful water supply, will result in increased uptake compared to conditions resulting in a high stomatal resistance, namely dark and drought.

2.2. Carbon

Once CO$_2$ is within the stomata it is assimilated and it is possible that metabolic factors play a role in uptake (Collins, 1994). The process of CO$_2$ uptake by vegetation, namely photosynthesis, is well documented and the reader is referred to general plant physiology texts, e.g. (Salisbury & Ross, 1992; Jones, 1992). It has been reported that CO$_2$ uptake as a result of photosynthesis is 1000 times more important than washout in removing $^{14}$CO$_2$ from the atmosphere (Chamberlain, 1988). As washout is the main pathway to soils below a canopy, it is unlikely that root uptake from soils plays a significant role in plant uptake of $^{14}$CO$_2$.

2.3. Sulphur

Whilst some studies have demonstrated the emission of COS by plant/soil systems (Rennenberg et al., 1990) the majority of studies suggest that COS can be assimilated, metabolised and incorporated into plant material and soils (Taylor et al., 1983; Kluczewski, Brown & Bell, 1983; Brown et al., 1986; Mihalopoulos, Bonsang, Nguyen, Kanakidou & Belviso, 1989; Bartell, Hofmann, Kruezberg, Andreae, & Kesselmeier, 1993) with little subsequent loss of the assimilated sulphur (0.5%) (Kluczewski et al., 1985). As a result of declining anthropogenic emissions of sulphur, plants may be using alternative atmospheric sulphur compounds as a source of sulphur (Zhao & McGrath, 1997), and therefore may be increasing their uptake of COS, and hence CO$_{35}$S. Once inside the plant, COS is assimilated by carbonic anhydrase, which is believed to be allied to the primary photosynthetic enzyme ribulose bisphosphate carboxylase oxygenase. Carbonic anhydrase has a specificity for COS which is 1000 fold greater than that for CO$_2$ (Protoschill Krebs, Withelm & Kesselmeier, 1996) and results in the reaction COS + H$_2$O $\rightarrow$ CO$_2$ + H$_2$S, with the resulting H$_2$S being assimilated into the amino acids methionine and cysteine (Kesselmeier, 1992).

Root uptake plays a smaller role than foliar uptake following gaseous deposition, due to the higher resistance of the air–soil–plant pathway. The deposition velocity of COS to soils has been calculated as $0.571 \times 10^{-6}$ and $3.06 \times 10^{-6}$ cm s$^{-1}$ for moist and dry soils, respectively, two orders of magnitude less than that for plants (Kluczewski et al., 1985). Therefore, even if all the labelled sulphur was transformed into a usable form such as sulphate, it is unlikely that this plays a significant role for COS uptake.

2.4. Tritium

As an isotope of hydrogen, tritium can enter essentially any organic compound. In the case of tritiated water, it will readily exchange with the stable water within the
plant. The HTO may then remain as tissue-free water tritium (TFWT), may be incorporated by exchange reactions between tissue water and exchangeable organic hydrogen or be assimilated into simple sugars by enzymatically catalysed reactions in which it replaces hydrogen to form OBT (organically bound tritium).

Several studies have been conducted on the flux of tritium within the plant. For example, it has been shown that tritium labelling and the subsequent dilution by stable hydrogen takes place at rapid rates during daylight, and at much reduced rates in the dark, thus suggesting that the main route of tritium uptake is via the stomata (Kline & Stewart, 1974). Uptake of HTO by tomato (Spencer, 1984), maize and barley (Garland & Ameen, 1979; Kim & Baumgartner, 1994) and wheat (Strack, Diabate, Muller & Raskob, 1995) has been demonstrated. In a study whereby potato and grape leaves were exposed to an atmosphere of HTO for 4 h, the tritium concentration of leaf water reached an apparent plateau after 45 min, which extended over the rest of the exposure period (Guenot & Belot, 1984). During the subsequent delabelling stage, tritium was lost from the leaf water at a rapid rate for several hours ($t_{0.5}$ 30 min), followed by a much slower rate ($t_{0.5}$ 30 h). Similarly, during an investigation in which maize was germinated and grown in an enclosure in which the soil water and the atmospheric vapour contained equilibrium concentrations of HTO, the maize was found to contain 95% of the environmental concentration of tritium (Garland & Ameen, 1979).

Dinner, Gorman and Spencer (1980) investigated the uptake coefficients for tomato and cucumber plants continually exposed to HTO vapour. Uptake coefficients in the range 0.4–0.8 were measured, with an average of 0.52 for ripe produce. The rate of uptake by foliage was rapid, but the rate of uptake by fruit itself was significantly lower, probably due to a slow rate of water turnover in the fruit.

The direct oxidation on plant surfaces of HT gas has been used as an explanation of elevated HTO concentrations found in forest floor litter (Murphy, 1984; Sweet & Murphy, 1984). However, specific investigations of the process found negligible HT conversion by vegetation such as tomato, corn and poplar (Dunstall, Ogram & Spencer, 1985). It is thought that the HT is converted to HTO as a result of bacterial action, as proposed by Bunnenberg et al. (1990). Deposition of HTO to soils is an order of magnitude lower than to vegetation (Bunnenberg et al., 1990), and also results in lower OBT levels than direct foliar uptake of HTO (Brudenell et al., 1997).

### 3. Allocation

#### 3.1. Carbon

Whilst work on $^{14}$CO$_2$ deposition to plants as an atmospheric pollutant is limited, $^{14}$CO$_2$ has been extensively used as a radiotracer in order to determine the allocation of carbon in vegetation. This has, therefore, provided a means of determining the path of $^{14}$CO$_2$ from the atmosphere to the plant, and the subsequent allocation within such vegetation.
Following assimilation of CO$_2$, carbon may remain localised, be transported in the xylem or phloem toward the leaf tip, transported in the phloem toward the stem, or be respired and therefore lost to the air (Troughton, Moorby & Currie, 1974). The tissues producing carbon assimilate are known as source tissues because they provide utilisable substances for transport to sink tissues where it is exploited or stored. Therefore, source tissues can be defined as net exporters of assimilate, whilst sink tissues are net importers. Carbon partitioning is the division of carbon among sinks which takes place by the regulation of import assimilates and accumulation or incorporation of carbon compounds in sink tissues (Geiger & Fondy, 1991). Essentially all plant organs will, at some point in development, act as sinks, whilst only some will act as source tissues (Ho, 1988). For instance, Thrower (1962) argued that in its life cycle, a leaf passes from a stage at which it imports assimilates, through a phase of simultaneous export and import to a stage at which export is predominant.

Fruit growth is part of the integrated growth of a plant, and it is sustained principally by the supply of current photosynthate (Ho, 1992). For assimilate partitioning, fruits are irreversible storage sinks, as the imported assimilate is either used for growth or stored as reserves and no net export occurs during the life of the organ (Ho, 1988). Whilst work on $^{14}$CO$_2$ uptake by fruits is limited, a large amount of information is available on $^{14}$CO$_2$ movement to similar organs such as root crops and cereal grains, which also act as irreversible storage sinks (Wardlaw, 1990).

Although phloem transport can occur over large distances, a sink is generally supplied with photosynthate from a nearby source, with the association between specific sources and sinks continuously changing (Wardlaw, 1968). Studies on the distribution of $^{14}$CO$_2$ in jute have shown that the top leaves supply carbon to the apical bud and young growing leaves, the centrally placed leaves supply carbon to the stem, and the basal leaves supply carbon to the lower stem and roots (Wardlaw, 1990). In a study on apple trees it was shown that the photosynthates from leaves of fruit-bearing apple spurs are almost exclusively transferred to the fruits on the same spur (Hansen, 1967).

In some cases the carbon accumulating in the fruits is self-generated. For instance, it has been suggested that the outer cell layers of the apple fruit photosynthesise, but only at one-tenth of the rate of leaves (Mooney, 1972). Many unripe fruits have a green colouration due to chlorophyll in the outer peel or tissues, and during the early stages of development, the fruit epidermis contains stomata and well-developed chloroplasts, the engines of photosynthesis. As the fruit develops, the photosynthetic pigments decline with ageing and ripening, reducing the photosynthetic capacity of the fruit (Schaedle, 1975).

A recent study involving translocation and localization of $^{14}$C-labelled assimilates in grapefruit showed that during the most rapid period of fruit growth, approximately 58% of the total $^{14}$C-labelled photosynthate was transported from the source leaf into the fruit within 24 h, with this percentage decreasing as the fruit developed. Distribution of this assimilate changed during fruit development, with the initial photosynthate contributing partly to the flavedo (outer 2–3 mm of pigmented peel) and later the juice sacs becoming the dominant sink. It was also shown that the flavedo appears to be able to fix CO$_2$ either from the external environment or from endogenous
respiration, and can therefore contribute to net photosynthesis. Whilst most of the photosynthate remained in the flavedo, it was shown that substantial inward diffusion of CO₂ can apparently take place in these fruit. Therefore, leaf photosynthates primarily supplied the juice tissues, whilst fruit photosynthates remained mostly in the peel (Yen & Koch, 1990). However, in developing apricots, the opposite is observed, with ^14CO₂ applied to the surface of the fruit being distributed uniformly throughout the fruit tissue (Bollard, 1970).

Studies conducted on peach and apple trees have demonstrated that leaves showed greater net photosynthesis during periods of rapid fruit growth when sink demand would probably be greatest. Therefore, during fruit growth, there is likely to be a higher rate of uptake of ^14CO₂. It has also been demonstrated that any effect of fruit on net photosynthesis was dependent on the stage of fruit development (Roper, Keller, Loeschger & Rom, 1988; Heim, Landsberg, Watson & Brain, 1979).

3.2. Sulphur

Bonas, Schmitz, Rennenberg and Bergmann (1982) proposed that “the distribution of ^35S follows source–sink relationships for reduced sulphur compounds and sulphate. The directionality is probably not determined solely by concentration gradients for sulphur compounds, but by an overall concentration gradient for ions, carbon, nitrogen and sulphur assimilates”, thus suggesting that sulphur allocation is similar to carbon allocation. This proposal is supported by Collins and Bell (1996), who found that 0, 5 and 20% of assimilated CO^35S was recovered from the pods of dwarf beans 24 h after exposure when exposed at the early, middle or late season, this correlated with the growth rate of these organs at the time of exposure. In a study whereby a mature source leaf of Ricinus communis L. was fed [^35S]sulphate, and phloem exudate collected from a site 34 cm below the source leaf, ^35S was detected at the collection point within 35 min from the start of the experiment, thus suggesting a translocation rate of at least 60 cm h⁻¹ (Bonas et al., 1982).

3.3. Tritium

Murphy (1993) stated that “The transport and cycling of tritium in the environment can be understood in terms of the role of hydrogen in the environment. Physical and chemical isotopic effects, while present in some transport mechanisms and chemical reactions, are not important factors in environmental tritium dynamics. The lack of significant isotopic effects or biomagnification means that tritium transport and cycling in the environment can be predicted on the basis of the transport processes, hydrogen content, and chemical transformation of hydrogen and its compounds in the environment”.

There is only a limited coverage in the literature on tritium and tritiated water behaviour in fruit crops: examples include work on wine grape (Vitis vinifera) (Belot, Gauthier, Camus, Caput & Bourdeau, 1979; Kirchmann & Fagniart, 1982; Guenot and Belot, 1984), sweet orange (Citrus sinensis) and olive (Olea europaea) (Kirchmann and Fagniart, 1982), tomato (Spencer, 1984), the wild shrub bristly blackberry (Rubus
setosus) (Edlund, 1988), strawberry (Fragaria sp.) and the shrub northern dewberry (Rubus flagellaris L.) (Spencer et al., 1988), plums, apples, peaches (Murphy, Bauer & Zeigler, 1992) and fruits of cherry tomato (Amano, Atarashi, Noguchi, Yokoyama, Ichimasa & Itchimasa, 1995). It should be noted, however, that although these authors report on HTO uptake by fruit crops, there is very little information available for allocation to the edible portion of the crop. A series of studies with respect to chronic HT release includes data from three species of plant: cherry tomato (Lycopersicon esculentum), radish (Raphanus sativus) and chinese mustard (Brassica campestris) (Davis, Galeriu, Spencer & Amiro, 1995a; Davis et al., 1995b; Noguchi, Yakoyama, Kinouchi, Murata, Amano & Atarashi, 1995; Amano et al., 1995). In the latter study, trace amounts of HT were released continuously to the surface atmosphere over a 12-day period. In the case of cherry tomato and radish, both leaf samples and samples of the edible portion of the plant were taken. The TFWT concentration for radish was shown to be similar for both the leaf and the root, whilst in tomato, the leaf HTO concentration is higher than that of the fruit. This is presumably due to the fruit having only indirect access to HTO from the leaf or root, whilst the radish ‘root’ has direct access from the soil. However, the OBT concentration for both radish root and tomato fruit was found to be considerably lower than that of the leaf. The specific activity of OBT in the plants gradually increased from the start of exposure. However, as the OBT concentration did not attain steady state over the exposure period, it is not possible to predict the ultimate OBT distribution (Amano et al., 1995).

$R$ values (stable OBT/free water HTO) were calculated from the data of Koranda and Martin (1973) for half-hour vapour exposures of sunflower. After 7 h, upper foliage had $R$ values between 0.56 and 2.17, whilst stem values ranged between 0.1 and 12.2, the latter was presumed to be due to translocated photosynthate. Guenot and Belot (1984) concluded that, as the loss rate of non-exchangeable OBT is much slower than for HTO, ratios of tissue-bound $^3$H to TFWT may exceed unity very soon after exposure. Spencer (1984) found $R$ values of up to 1.92 in the green fruits of tomato plants exposed to atmospheric HTO. The results show that the fruits have little exchange with atmospheric moisture, hence they maintain a low TFWT value; however the fruits incorporate photosynthetic products labelled with tritium in non-exchangeable positions.

4. Loss

It is important to emphasise the large differences in the rates of loss between the three radionuclides. For carbon the loss can be ascribed to respiration of carbon dioxide, this can be of the order of 25–50% of photosynthetic assimilation depending on environmental conditions (McCree, 1974), the remainder becomes fixed in plant material and will only be respired if there is re-allocation of this material such as the use of stem reserves in the later stages of grain growth in wheat. The loss of $^{14}$C activity following exposure of whole plants of bean, cabbage and wheat was best fitted by a single exponential with a biological half-time of 43 d (Collins & Bell, 2000). The same function fitted to the data of Hume and Criswell (1973) for
soybean gave a half-life of 50 d, while the value for a range of grain legumes reported 54% loss after 55 d (Pandey, Saxenam Jalubarme, Singh & Prasad, 1976) and a range of wheat cultivars lost 45% of activity 22 d after exposure (Gent & Kiyomoto, 1989).

Data for the loss of $^{35}$S from crops are limited. Collins and Bell (1996) reported that there was no significant loss of $^{35}$S post deposition and that all losses could be accounted for by radioactive decay ($t_{1/2} = 87.5$ d) following exposure to either CO $^{35}$S or H$_2$ $^{35}$S. Thus the known flux of sulphur-containing gases away from crops (Carrol, Heidt, Cicerone & Prin, 1986; Goldan, Kuster, Albrittin & Fehsenfield, 1987; Bartell et al., 1993) is proposed to occur from labile pools of sulphur. Therefore once the $^{35}$S is fixed post deposition it is not available to these pools and hence losses are minimal.

This contrasts with the more rapid loss following HTO exposure where a biphasic pattern was observed with an initial half-life of 30 min, due to vapour exchange and transpiration, and a $t_{1/2}$ of 30 h for a second component from tissue water of potato and grape leaves, thought to have been labelled by phloem translocation (Guenot & Belot, 1984).

An appraisal of the literature data leads to the conclusion that the methods used to apply tritiated water to an experimental plant lead to a bias in derived parameters and precludes the application of some literature data for use in the dynamics of short-term atmospheric releases of tritium. The literature includes studies involving the application of HTO by pipette into bore holes into the trunks of tropical trees (Kline, Martin, Gordon & Koranda, 1970), and via the nutrient solution into 1–2 yr old Sour Orange seedlings (Citrus aurantium) (Mantell, Monselise & Loldschmidt, 1979).

The range of techniques, experimental designs and aims, as well as species used in HTO tracer studies is reflected in the range of mean residence times (rate of water turnover in the plant) of tritium which varies widely for forest trees, from 23 h to 11 d and which for citrus leaves has been reported to be about 30 h (Mantell et al., 1979). In the case of herbaceous plants, the value is much lower. Many of the above studies were designed to measure transpirational losses using HTO as a tracer or were concerned with the soil–plant pathway. Diabate and Strack (1993) stated that “Most of the total hydrogen contained in green plants is associated with the tissue water fraction, which represents some 90% of the fresh plant material. The relative amount of hydrogen in water is $\sim 11\%$ while that in organic material ranges from 6–7%. If some 30% of organically bound hydrogen is assumed to be exchangeable, the magnitude of the non-exchangeable OBT compartment can be estimated as only 3% of the total hydrogen in fresh plant material”.

A time course of formation of non-exchangeable OBT indicates that non-exchangeable OBT formation increases over a 24 h period, with a marked decrease after 3 d indicating a dynamic turnover of OBT. This observation is supported by the non-exchangeable $^3$H in the organic matter of leaves of potato (Solanum tuberosa) and wine grape (Vitis vinifera) (Guenot & Belot, 1984) with about 50% of the initial concentration of tracer lost during the first few hours. The remaining activity decreases with a half-life of approximately 80 h.

Non-photosynthetic assimilation in the dark is more important for tritium than for $^{14}$C (Diabate & Strack, 1993). The production rates of OBT have been represented by
linear equations after 75 h, because HTO concentration in the surface environment almost attained an equilibrium state within 3 days of the initiation of an HT release (Amano et al., 1995). The production rates differed with the different species (Chinese mustard, tomato fruits and radish roots) and OBT concentrations were still rising at the end of a 12 d HT release, suggesting that the system was not at steady state with respect to OBT. A study on the translocation of tritiated assimilates into potato and wheat grains has been reported (Diabate & Strack, 1992).

The effect of the method of application of HTO has a profound effect on the levels of both TFWT and hence non-exchangeable OBT. Uptake of HTO vapour via the soil pathway only results in very low OBT levels (Brudenell et al., 1997). Uptake via vapour phase into plant leaves results in moderate levels of OBT, whilst soil-applied tritiated water results in OBT levels an order of magnitude higher (Brudenell, unpublished). This finding is in agreement with work on vegetables that had been sprayed with HTO (Kirchmann & Fagniart, 1982). The allocation of tritium within the plant will follow that of water in the transpiration stream. When fixed as OBT the tritium is probably allocated along with $^{35}$S and $^{14}$C by source/sink relationships. There are however very few data to illustrate this.

5. Conceptual model

The modelling of radionuclides following gaseous deposition is outlined in Fig. 2. There is deposition to the leaves that is dependent on the $V_g$, time of exposure and air concentration of the nuclide of concern. The total leaf area for deposition can be derived from the literature on leaf area indices (area of leaf per unit area of ground). There will then be allocation to stem, root and fruit (if present). Allocation to the fruits would be dependent on their rate of growth at the time of exposure. In general, most fruits, e.g. apple, strawberry and tomato, can be represented by a simple sigmoid growth curve, starting with an exponential increase in size, and then slowing in a sigmoid fashion. However, a second group of fruits has a more complicated growth curve, involving two periods of growth increase with a period of slow or suspended growth in between e.g. peach, apricot, grape and currant (Moore, Clark, Stern & Vodopich, 1995). The losses of the activity could be described by simple loss rates, equivalent to radioactive decay for $^{35}$S, a $t_{1/2}$ of 50 d for $^{14}$C and a biphasic loss for tritium with 30 min half time initially followed by 7 d to account for both HTO and OBT.

6. Conclusions

Mechanisms for the uptake of CO$^{35}$S, HTO and $^{14}$CO$_2$ are well understood and their deposition velocities quantified. There is also a reasonable body of work detailing the translocation of $^{14}$C once in the crop, but much less for $^{35}$S and $^3$H, which are considered to follow source sink relationships. The loss rates of the three radionuclides show large differences with tritium lost rapidly in the form of HTO but
Fig. 2. Conceptual model of gaseous uptake by crops.

retained longer when converted to OBT. Losses of $^{14}$C are less and those of sulphur are minimal post fixation. When fruit crops alone are considered, the quantity of information is further reduced, but predictions on possible behaviour of these radionuclide species can be made from current knowledge.

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