



Tritium in plants: A review of current knowledge

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ABSTRACT

The aim of this paper is to provide an overview of the literature regarding the fate of tritium in plants. At first, some prerequisites about the sources, chemical and physical characteristics, biological effects and potential risks associated to tritium are presented. The general scheme of tritium transport and cycling in the environment, the different tritium fractions in biological organisms and the corresponding techniques for sampling and measurement are discussed. Then, the main processes of uptake, incorporation and conversion of tritium into plants are described, based on experimental data. As tritium incorporated into the organic fraction may be the major contributor to the dose of radiation received following its ingestion by man, the biological mechanisms and kinetics of organically bound tritium (OBT) production are a major focus. Some predictive models of short-term or chronic exposure are described. Finally, the pending questions, concerning the fate of tritium in plants, are tackled.

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1. Introduction

The global inventory of tritium, the only radioactive isotope of hydrogen, is too low to present a radiological risk on a worldwide scale, and natural tritium production remains far higher than man-made sources. However, release of tritium from punctual sources is likely to increase in the near future with the development of fusion technologies. Because of the potential health hazard associated to tritium (Rohwer and Wilcox, 1976; Straume, 1993), local evaluations of the radiological risks are required (Okada and Momoshima, 1993; Gulden and Raskob, 2005).

The radiological impact of tritium results from the combination of the characteristics and the behaviour of the radionuclide. Some parameters enhance the radiotoxicity of tritium: for example, its radioactive half-life relatively long (12.35 years), its uptake by humans is easy and as an isotope of hydrogen, it has a high biological

importance (Rohwer and Wilcox, 1976). On the other hand, other features lead tritium to be considered it as a nuclide of low radiotoxicity. Indeed, the low-energy beta particle emitted by tritium has a maximum range in water or tissue of 6 μm , which prevents it from being an external exposure hazard. As a consequence, tritium is associated to a radiological risk only if absorbed by the human body, notably after ingestion of tritiated organic molecules. Moreover, its biological half-life in the water pool of living organisms is very short (10 days).

The physical and chemical properties of tritium are almost the same as hydrogen. Since hydrogen is the most abundant element on earth in terms of number of atoms and one of the main elements in the biosphere, especially as a constituent of water, understanding the behaviour of tritium in the different ecological compartments (e.g. atmosphere, soil, water, microflora, vegetation, fauna) is necessary to estimate the risk for man and for the environment. Tritium's fate in vegetation is of particular interest because photosynthesis is the necessary first step for the production of organic matter which then moves through the whole environment, until its potential uptake by humans (Elwood, 1971).

This paper summarizes the information available about the sources, transport, transfer, uptake and incorporation of tritium in plant systems, with special attention to plants involved in the human diet. The knowledge resulting from experimental data and the predictive models mostly used are described in detail. The principal gaps in current knowledge about the fate of tritium in plants are emphasized; they concern experimental data on chronic and acute exposure, the effects of low-level radiation and the biological aspects of tritium uptake by plants.

Abbreviations: BIOMASS, BIOSphere Modeling and ASSESSment; BIOMOVs, BIOSphere MODEL Validation Study; EMRAS, environmental modeling for radiation safety; HWR, heavy water reactor; IAEA, International Atomic Energy Agency; ICRP, International Commission on Radiological Protection; ITER, International Thermonuclear Experimental Reactor; LET, low energy transfer; OBT, organically bound tritium; PAR, photosynthetically active radiation; PWR, pressurized water reactor; RBE, relative biological effectiveness; SA, specific activity; SAR, specific activity ratio; TFWT, tissue free water tritium; TLI, translocation index; UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation; VAMP, VALIDation of Model Predictions; WHO, World Health Organization.

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Moreover, the absence of a standard concerning organically bound tritium (OBT) prevents the validation of any analytical methods for OBT measurement and comparison between experimental or modeled data. Additional experimental data and characterization of standard reference materials, especially for environmental concentration ranges, are needed.

2. Prerequisites about tritium

It is not the purpose of this paper to review the large volume of literature that describes the processes of tritium production or transport between the different compartments of the environment. However, this section deals with the key mechanisms and definitions for understanding tritium transfer and its fate in plants. Extensive literature is available for more details about tritium origins, transport and cycling in the environment (Elwood, 1971; Bogen and Welford, 1976; Eisenbud et al., 1978; Momoshima et al., 1991; Murphy, 1993; Okada and Momoshima, 1993).

2.1. Sources of tritium

Tritium in the environment has three sources: natural production, release from atmospheric weapon tests, and routine or accidental releases from the nuclear industry (Okada and Momoshima, 1993; Belot et al., 1996). Totally, the world burden of tritium is estimated to be approximately 1300 PBq (3.5 kg), which is close to the natural equilibrium level (Belot et al., 1996; Hisamatsu et al., 1998; UNSCEAR, 2000).

The reader should keep in mind that tritium amounts are expressed by their mass (grams) or by their activity (Bq). One gram of tritium corresponds to 3.58×10^{14} Bq (i.e. 358 TBq or 0.358 PBq).

2.1.1. Tritium in nature

The natural production of tritium mainly arises from reactions induced by cosmic rays in the upper atmosphere (Kaufman and Libby, 1954; Okada and Momoshima, 1993). Most of the time it results from the interaction between a fast neutron and atmospheric nitrogen following (1). Another, very small, fraction of natural tritium is produced by neutron capture by ${}^6\text{Li}$ in the earth's crust (Kaufman and Libby, 1954; Okada and Momoshima, 1993). The overall natural production of tritium ranges between 50 and 70 PBq (0.15–0.20 kg) in a year.



Activities measured before the nuclear bomb tests (in the 1950s) were about 0.6 Bq L^{-1} in precipitation, $0.3\text{--}0.8 \text{ Bq L}^{-1}$ in rivers and lower than 0.1 Bq L^{-1} in seawater (Momoshima et al., 1991). Tritium activities in rain reached 150 Bq L^{-1} in 1963 in the Northern hemisphere because of atmospheric bomb tests. At present, HTO activities in nature are close to those recorded in the pre-nuclear period; 1 Bq L^{-1} in precipitation, less than 10 Bq L^{-1} in rivers, $1\text{--}10 \text{ Bq L}^{-1}$ in aquifers, less than 0.3 Bq L^{-1} in seawater, $2\text{--}4 \text{ Bq L}^{-1}$ in the ice of the poles (Belot et al., 1996).

2.1.2. Tritium released by atmospheric thermonuclear bomb tests

The man-made tritium released into the environment comes from several sources. The most important one is the atmospheric weapon tests performed between 1952 and 1963. During this period, about 560 kg of tritium (240 000 PBq) were released into the atmosphere in both hemispheres. Because of radioactive half-life (12.35 years), the tritium currently remaining from the nuclear tests was estimated to be 40 kg in 2008, distributed between oceans, continental water and the atmosphere (Guétat et al., 2008a, b).

2.1.3. Tritium released by nuclear plants

The main sources of tritium released by man are linked to the nuclear power cycle: nuclear power stations, nuclear fuel reprocessing plants or tritium production plants (Okada and Momoshima, 1993). Most significant leakages are due to reprocessing plants; releases are essentially liquid, and reach several PBq annually (for example 10 PBq a year for the reprocessing plant "La Hague" in France). In Heavy Water Reactors HWR (for example the Canadian reactors CANDU), airborne effluent ranges from 0.1 to 1 PBq and liquid releases from 0.1 to 0.5 PBq a year. The nuclear thermal power stations using light water (pressurized water reactors, PWR) have rather low discharges: atmospheric releases range from 0.3 to 3 TBq and liquid releases are about 20 TBq a year. Moreover, some other industries are "small users" of tritium, for medical and R&D purposes or tritium light facilities. All these kinds of devices produce few releases (some PBq per year).

Total industrial releases (HT gas and heavy water HTO) are currently about $0.06 \text{ kg year}^{-1}$ (Belot et al., 1996); while natural production of tritiated water ranges from 0.15 to $0.20 \text{ kg year}^{-1}$.

2.1.4. Future usage of tritium

In the near future, some applications could lead to new sources of tritium. The future usage of tritium mainly concerns nuclear fusion facilities (like the International Thermonuclear Experimental Reactor, ITER). The quantity of tritium used by ITER would amount to 1.5 kg year^{-1} ; the associated release in the atmosphere could be significant. On a much smaller scale, high-power lasers (like the Laser MegaJoule from the French atomic agency, CEA) could use a few milligrams of tritium per year (Guétat et al., 2008a, b).

2.2. Chemical and physical characteristics of tritium

Tritium is the only radioactive isotope of hydrogen (atomic number: 1; atomic weight: 3). The physical and chemical properties of tritium are very close to those of hydrogen (Eisenbud et al., 1978). Tritium occurs mainly in the form of tritiated water, but other chemical species of tritium are naturally present, principally as tritiated molecular hydrogen (HT) and tritiated methane (CH_3T). Transport of these different chemical species of tritium in the environment is related to physical, chemical and biological processes. Physical processes are bulk transport (tritium moves with the bulk flow) and diffusional transport (tritium follows a gradient of concentration from higher to lower level). Reactions and state changes of chemical species are chemical processes (Murphy, 1993). Biological processes such as photosynthesis, respiration or digestion concern the transfer of tritiated molecules through biological organisms.

The mass difference in the isotopes of hydrogen (protium, deuterium and tritium) leads to slight isotopic differences in chemical and physical processes. Theoretically, slower chemical reactions occur with the heavier isotopes. However, under environmental conditions, the tritium pool is very low compared to that of hydrogen. For instance, 0.6 Bq L^{-1} in precipitations represents about 10^{-17} kg of ${}^3\text{H}$ per kg of H_2O . Thus, the influence of HTO on the evaporation rate and on most chemical properties of the aqueous solution is negligible (Eisenbud et al., 1978; McFarlane et al., 1979b). In other words, there are no apparent isotopic effects or biomagnification in natural processes (Elwood, 1971; Eisenbud et al., 1978; Murphy, 1993; Belot et al., 1996); tritium transport and cycling in the environment can be considered as identical to that of hydrogen and therefore, of water (Elwood, 1971; McFarlane et al., 1979b; Momoshima et al., 1991; Murphy, 1993; Okada and Momoshima, 1993).

2.3. Biological effects and risks related to tritium

2.3.1. Biological effects of tritium

The most appropriate source of information for estimating the risks for human health from tritium exposure is epidemiological studies. A large number of such studies involving nuclear workers, members of the public and their children have been reviewed (Little and Wakeford, 2008). However, the lack of tritium-specific dose data and the small numbers of cases prevent any inferences from being made on health effects specifically related to tritium exposure (Little and Wakeford, 2008). Because few data are available about tritium-induced health injury in humans, its radiological effects have been evaluated from observations on exposed cells or animals.

In our opinion, there is no convincing data available about the biological effects of tritium in plants. No noticeable effect on biomass production has been observed during experimental tritium exposure of various vegetables (Choi et al., 2000). However, we can assume that DNA mutations and cell death may occur for very high-level tritium exposure in the same way as in animals.

2.3.2. The relative biological effectiveness and radiation weighting factor of tritium

The results of animal experiments are not directly transposable to humans. Indeed, the biological effectiveness of the weak beta emission for different irradiation conditions has to be considered to assess tritium radiotoxicity for human beings (Rohwer and Wilcox, 1976). The relative biological effectiveness (RBE) of tritium is the ratio between the dose absorbed due to tritium beta radiation and the dose absorbed due to a standard radiation (X or gamma) to produce the same specified effect. RBE data are used by ICRP, together with biophysical considerations, as the basis for specifying radiation weighting factors. A value of one is applied to all Low Energy Transfer radiation (LET), despite observed RBE of 3–4 with gamma rays (Melintescu et al., 2007; Galeriu et al., 2008). The reasonable justification is that in general the greater values may not apply to cancer induction in humans (ICRP, 2007) and undue precision may be implied by the use of different weighting factors w_R for different low LET radiations (Harrison, 2007).

2.3.3. Ways of exposure to tritium for humans

Inhalation, ingestion and skin exposure are the three main routes of exposure to tritium for mankind. However, it is important to notice that tritium is a pure beta emitter of low energy; the maximum energy of electrons emitted from tritium decay is 18.6 keV. The corresponding maximum track length is 6 μm in water or biological tissue and the thickness of the epidermis and dermis of human skin is 20–100 μm and 1–3 mm, respectively. As a consequence, tritium could inflict damage on humans only when it is present inside the body (Okada and Momoshima, 1993). Fortunately, the turnover of tritium in the human body is very rapid; even in the case of absorption, tritiated water, which represents 90% of the tritium absorbed, is expelled with a biological half-life of approximately 10 days. Organic tritium (10% remaining) maintains tritium within the body for a half-life of approximately 30 days. Finally a small fraction of organic tritium remains in the fat tissues or collagen for approximately 450 days (Okada and Momoshima, 1993).

2.3.4. Tritium dosimetry for humans

Consequently, the radiotoxicity of tritium is relatively low: the dose coefficients per unit of incorporation (the effective dose per built-in Becquerel) have been evaluated at 1.8×10^{-15} Sv Bq⁻¹ for HT (inhalation), 1.8×10^{-11} Sv Bq⁻¹ for HTO (ingestion or inhalation) and 4.2×10^{-11} Sv Bq⁻¹ for OBT (ingestion) for an adult person. The occurrence of a cancer due to chronic exposure to tritium

cannot be excluded, although epidemiological data are not yet sufficient to quantify the risk. As for other radionuclides, the theoretical risk of death by cancer due to tritium incorporation has been calculated to be 6.5×10^{-2} per Sv incorporated (Straume, 1993).

The World Health Organization (WHO) places the limit at an annual effective dose of 0.1 mSv, corresponding to the daily consumption of tritiated water at a concentration of 10000 Bq L⁻¹ during a year. On the other hand, European Directive 98/83 fixed an investigation threshold of 100 Bq L⁻¹ for tritium radioactivity in the water supply. Besides, some researchers in the international tritium community consider that health effects of tritium are not sufficiently understood and that further studies are needed to decrease the uncertainty of radiological assessments (Little and Wakeford, 2008).

2.4. Global scheme of tritium transport and transfer in the environment

2.4.1. Tritium in the biogeological cycle of water

As an isotope of hydrogen, tritium follows the biogeological cycle of water. The global scheme of this cycle in a temperate country is summed up in Fig. 1. About 13% of the precipitations and condensations, and thus of deposited tritium, are re-emitted in the atmosphere by evapotranspiration. More than a third is firstly incorporated into soil water and then absorbed and transpired by plants. It is very important to note that more than 50% of water is directly or indirectly evaporated by the plants after falling from the atmosphere. Consequently, vegetation plays a crucial role in the water exchanges occurring in terrestrial ecosystems. Vegetation is very likely a key factor governing tritium exchanges inside these ecosystems. Almost 40% of precipitations form runoff or are incorporated into the subsurface waters (rivers). Finally, a small remaining fraction (about 10%) penetrates the soil and infiltrates into the deeper layers of groundwater under the influence of diffusion, gravity and capillarity (Murphy, 1993). Then, tritium follows the movement of underground water; its residence time varies according to the characteristics of the environment. The oceans are the major global reservoir of hydrogen on earth and thus, of tritium. Because of their enormous capacity of dilution for tritium (Murphy, 1993; Belot et al., 1996), most of the tritium released into the environment by human activities finally resides in the oceans.

2.4.2. Atmospheric circulation and deposition of tritium

The main chemical forms of tritium in the atmosphere are, in order of importance: tritiated water vapour (HTO), tritium gas (HT) and tritiated methane. Because tritium levels in the atmosphere are not high enough to imply density effects, the tritium follows the air flows and is diluted by atmospheric dispersion like any other trace element (Belot et al., 1996). The physical mechanisms involved have been largely described in the literature (Pasquill and Smith, 1988).

The atmospheric processes of tritium removal from the atmosphere are classified as wet deposition or dry deposition according to whether precipitations are the cause of the deposition on the soil or not. Dry deposition occurs on contact with tritiated air via processes of collection through leaf epidermis or the ground. Wet deposition results from scrubbing of the air by rain, snow and fog. Due to its low solubility, tritium in gaseous form HT and tritiated methane are rather concerned by dry deposition whereas the tritiated water vapour could be either deposited directly on the soil (dry deposition) or contained in the raindrops (wet deposition).

The deposition velocity of HTO to soil, i.e. the amount of deposition per unit area divided by the concentration in the atmosphere, typically depends on the texture and the surface soil water content. The deposition velocity of HTO to plants is dependent on plant characteristics such as the leaf area index and the stomatal resistance. For both soil and plants, the deposition velocities of HTO ranges

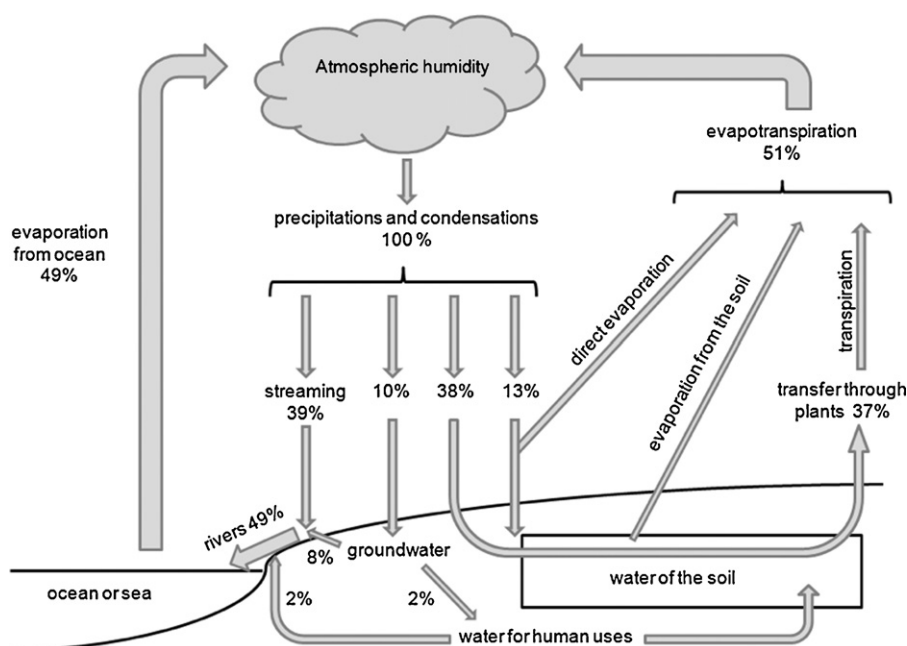


Fig. 1. Cycle of water corresponding to a temperate climate. This scheme corresponds to a moderately wooded, but very urbanized plus cultivated land (redrawn after Keller, 1980).

from 0.001 to 0.1 m s^{-1} as a function of atmospheric stability and surface properties (Eisenbud et al., 1978; Galeriu et al., 2008). HT deposition velocity to soil is estimated to be 10 to 100 times lower than that of HTO (Murphy, 1993; Koarashi et al., 2001). Its value depends on the meteorological conditions, the soil properties and the amount of soil bacteria.

2.4.3. Movement of tritium in soil and groundwater

In the event of continuous tritium deposition on the ground, an exponential profile of tritium concentration could be formed in the exposed soils (Belot et al., 1996). The characteristic penetration length for HT was determined to be only 2.3 cm because the deposited tritiated hydrogen on the soil is quickly oxidized and transformed into HTO due to microorganisms such as bacteria, protozoan microorganisms, mosses (McFarlane et al., 1978; Garland and Cox, 1980; Sweet and Murphy, 1981). As a consequence, all the tritium deposited on the ground (HT and HTO) finally joins the water pool of the soil and participates in every aspect of water circulation, such as evaporation, transport to ground water, and uptake and transpiration by plants (Eisenbud et al., 1978; Papke and Foerstel, 1991; Murphy, 1993).

Most of the tritiated water in the soil is reemitted into the atmosphere. The reemission rate of tritiated water in steady-state conditions was evaluated at approximately 1% per hour (Belot et al., 1996). During the first hours following a deposition of tritium (HT) the reemission rate is high. Täschner and Bunnenberg (1997) have evaluated the rate at 28% h^{-1} in the first hour. After a single and short tritium exposure, the activity of the radionuclide in the soil decays exponentially with a half-life of some days. Kirchmann et al. found half-lives of 1–2 days in summer and 5 days in winter (Kirchmann et al., 1971; Belot, 1986), whereas Koranda and Martin reported about 4 days (Koranda and Martin, 1971).

In the humic layer, part of tritium as HTO is used to build organic matter during normal biomass production of soil microorganisms. In an experiment conducted with five different soils (brown soil, para brown soil, rendzina), Papke and Foerstel (1991) determined an average rate of OBT formation related to the mean HTO activity in the soil. This rate ranged from 0.03% to 0.05% per week, but rose to 0.35% per week with the addition of a substrate (glucose) to the

soil. OBT formed in soil was found to have a mean residence time of 5 years (Diabaté and Strack, 1993), before being metabolized as HTO.

Into the soil, liquid tritiated water is carried by bulk flow in response to gravity and capillarity forces. Tritiated water can also move by vapour diffusion, following a concentration gradient from higher to lower levels (Murphy, 1993). Infiltration of tritiated water depends on several factors such as the type of soil (clay, loam, sand), the state of the soil (cultivated or not), the water content and the organic matter content. Almost all of the tritium entering the groundwater (the ground extending from below the rooting depth of the vegetation to bedrock) is in the form of liquid tritiated water and is transported by bulk flow. In general, the velocity of groundwater flow ranges from about 1.5 m year^{-1} to about 1.5 m day^{-1} (Eisenbud et al., 1978; Murphy, 1993).

2.5. Tritium pools in biological organisms

Tritiated water is easily incorporated into biological organisms as tissue free water tritium (TFWT). Considering plants, water contents typically represent some 80–95% of the fresh mass of the living organs, but this percentage strongly depends on the species and on the stage of development (McFarlane et al., 1979b). It averages 85% in fruit, 75% in tubers and can be lower than 10% in some organs like seeds (IAEA, 2007b). Once got in the water pool, tritium may label the organic matter as organically bound tritium through metabolic processes, especially photosynthesis in the case of plants. Usually, tritiated organic matter is divided into several pools according to the nature of the chemical bonds involving tritium (labile or not). Usually, non-exchangeable OBT is defined as non-labile tritium bound to carbon atoms unlike labile exchangeable OBT bound to sulphur, nitrogen or oxygen atoms (Belot, 1986; Diabaté and Strack, 1993; Baglan et al., 2005). Table 1 presents a summary of the different pools of tritium in plants.

Exchangeable organic tritium is in equilibrium with the plant water pool and behaves globally like the TFWT fraction, with the same half-time in vegetation (Guenot and Belot, 1984; Galeriu et al., 2008). Conversely, because C–H (and so C–T) bonds are stable, non-exchangeable OBT is voided much more slowly than aqueous

Table 1

Definitions and respective importance of the different tritium pools in plants. Tissue free water tritium (TFWT) is the tritium present in the water pool of the plants. The tritium incorporated into the organic matter is called organically bound tritium (OBT).

100% T		
TFWT	Total OBT (5–10%)	
According to the species, 80–90%	Exchangeable OBT	Non-exchangeable OBT, buried tritium <1%
$\left\{ \begin{array}{l} \text{HT} \\ \text{HTO} \\ \text{T}^+ \\ \text{H}_2\text{TO}^+ \end{array} \right.$	$\left\{ \begin{array}{l} \text{C-T} \\ \text{S-T} \\ \text{N-T} \\ \text{P-T} \\ \dots \end{array} \right.$	C-T

or exchangeable OBT and therefore remains in the organisms for a long time (Guenot and Belot, 1984). Since OBT may be the main contributor to the human radiation dose in case of ingestion of contaminated food (Choi et al., 2002; Harrison, 2007), the OBT fraction is used for retrospective measures, biological survey, evaluation of atmosphere pollution, or dose estimation for humans near nuclear plants (McFarlane et al., 1979a; Kim and Han, 1999; Pointurier et al., 2004; Gulden and Raskob, 2005).

However, these definitions of exchangeable and non-exchangeable OBT have been the subject of controversy in recent years. Actually, tritium atoms covalently bonded inside biological macromolecules are exchanged very slowly because of their inaccessibility to cellular water and not because of the nature of the atoms they are bound to (Diabaté and Strack, 1993). Consequently, such “buried tritium” could behave as non-exchangeable OBT even if it is not necessarily bound to carbon (Baumgartner and Donhaerl, 2004). For the group of the Environmental Modeling for Radiation Safety (EMRAS) program in charge of tritium and ^{14}C questions (IAEA, 2007a), “OBT is carbon-bound and buried tritium formed in living systems through natural environmental or biological processes from HTO (or HT via HTO). Buried tritium is tritium that occupies exchangeable positions in large biomolecules in dry matter but that is not removed by rinsing with tritium-free water.”

Definitions of the exchangeable or not exchangeable OBT fractions may not be relevant from the biological point of view. It exists millions of organic molecules with their own transformation processes in vegetables, animals, microorganisms and man gastrointestinal tractus, and their own residence times. Indeed, tritium in initial water (HTO) can integrate organic molecules by biological reactions and may be incorporated into exchangeable or non-exchangeable positions. Tritium in an exchangeable status can also be located through a metabolic process into a non-exchangeable position, and vice versa. Definition of two classes will always be matter of discussions.

2.6. Tritium sampling and measurement in environmental samples

Levels of tritium activities and chemical forms could be very different according to the samples considered (releases, waste, biological samples...). As a consequence, the analytical technique to measure tritium has to be chosen considering the objectives of measurement. This paragraph deals only with sampling and measurement in environmental samples. The reader interested about tritium measurement in other types of samples can find extensive information in specialised literature (Wood et al., 1993; Belot et al., 1996).

2.6.1. Sampling of tritium in environmental samples

Tritiated water vapour can be sampled from the atmosphere by aspiration and bubbling of air through tritium-free water or desic-

cant (e.g. silica gel, calcium sulphate, molecular sieve) (Nakashima et al., 1981; Patton et al., 1997; Rosson et al., 2000; Uda et al., 2006). Atmospheric tritiated hydrogen and methane first have to be oxidized to be caught by bubbling systems as tritiated water.

As the most common form of tritium in the environment is tritiated water, samples of aqueous phases, such as rivers or seawater, can be analysed directly.

The methodology for the extraction and analysis of the different ^3H fractions (TFWT, exchangeable and non-exchangeable OBT) in soil and biological organisms has been largely described in the literature. Classical methods for TFWT extraction from samples are freeze-drying (Garland and Ameen, 1979; Kim and Baumgartner, 1988; Wood et al., 1993; Diabaté and Strack, 1997) or azeotropic distillation (Kim and Baumgartner, 1994). The total removal of TFWT was confirmed by weighing residual samples. Exchangeable OBT is removed by isotopic exchange. Several methods could be used. In most cases, dried samples are mixed with a sufficient volume of tritium-free water for 3 days. The isotopic exchange could also be carried out with tritium-free water vapour in an oven (Pointurier et al., 2004). Non-exchangeable OBT extraction methods involve a combustion apparatus in order to break the $-\text{C-H}$ and $-\text{C-T}$ bonds. The dehydrated samples are typically burnt under a flow of pure and dry oxygen, and resultant combustion water is condensed in a cold trap (Pointurier et al., 2003; Workman et al., 2005). Combustion with an oxygen bomb is also employed (Moghissi et al., 1975; Workman et al., 2005).

Dealing with tritium sampling, it is very important to keep in mind that tritium is very mobile which may lead to contamination from one sample to another. Besides, there is no standard procedure of sampling and preparation for tritiated samples yet. Some inter-laboratory comparisons of tritium measurement in environmental samples have underlined the discrepancies caused by the different used methods (Workman et al., 2005). In particular, the best way to extract or eliminate the exchangeable fraction of OBT divides the radioecologist community (Pointurier et al., 2003; Workman et al., 2005).

2.6.2. Measurement of tritium in environmental samples

Tritium activities in aqueous samples are directly measured by liquid scintillation spectrometry (Baglan et al., 2005; Workman et al., 2005). This technique consists in mixing tritiated water with a scintillating cocktail which transforms the β radiation into photons. The photons are then collected and measured outside the liquid. In optimal conditions of use, the detection limit of scintillation counters is below 1 Bq L^{-1} (Pointurier et al., 2003). However, this technique is sensitive to artefacts such as other radionuclides, organic compounds, chemiluminescence or static electricity (Guétat et al., 2008a).

When the tritium activities are low, electrolytic enrichment could be necessary. Mass spectrometry of ^3He is an alternative method to estimate low levels of tritium thanks to the radioactive decay (Kotzer et al., 1998). With this technique, the detection limit could be lowered to 0.01 Bq L^{-1} (Belot et al., 1996).

3. Uptake and incorporation of tritium in plants: knowledge from experimental data

Tritium uptake by vegetation, that is to say the transfer of tritium from the outside to the inside of plants, is the first step for the further incorporation of tritium into the organic fraction, in other words assimilation as OBT.

A reminder for the following is that C3 plants are so called because they make a three-carbon compound as the first stable product during photosynthesis while plants in which the CO₂ is first fixed into a compound containing four carbon atoms before entering the Calvin cycle of photosynthesis are called C4 plants.

3.1. Tritium transfer from environment to plants

3.1.1. Tritium transfer mechanisms

Tritium uptake depends on the chemical form of tritium considered. Actually, although tritium oxide (HTO) enters into the plants directly and easily as liquid water or water vapour, experimental studies have shown that tritium gas HT is not absorbed by vegetation due to its low solubility in water (Belot, 1986). The most common route of plant contamination by HT involves the oxidation into HTO in plant foliage (Belot, 1986; Spencer and Dunstall, 1986; Ichimasa et al., 1999) or in the soil (Stewart, 1971; McFarlane et al., 1978, 1979a; Bunnenberg et al., 1986; Spencer and Dunstall, 1986; Diabaté and Honig, 1988; Foerstel, 1988; Amano et al., 1995; Ichimasa et al., 1999). Indeed, the resulting reemission of tritiated water from the soil to the atmosphere by evapotranspiration could lead to the foliar uptake of HTO vapour (Foerstel et al., 1987; Belot et al., 1988; Okada and Momoshima, 1993; Amano et al., 1995). As the rate of HT to HTO conversion is high (for example 0.104–1.64 Bq per min and per gram of fresh matter (McFarlane, 1978) for potted lettuce plants *Lactuca sativa* L.), the overall mechanism of tritium transfer from environment to plants can be summarized as the HTO uptake.

Moreover, two main pathways for tritium uptake into plants have to be considered: the foliar uptake of tritium from the atmosphere, and the root uptake of tritium from the soil (Elwood, 1971; McFarlane et al., 1979b; IAEA, 2007b). The general scheme of tritium transport and cycling in the vegetation is presented on Fig. 2.

3.1.2. Tritium (HTO) transfer from the soil to plants

Tritium transfer from the soil to plants is associated with water uptake by the roots. Tritiated water from the soil follows a pathway similar to that of ordinary water through xylem sap, moving into the plants by bulk flow along a free-energy gradient (Murphy, 1993). The evaporation of water from the leaves maintains this gradient (Murphy, 1984). Plant exposure to tritium is strongly influenced by the concentration and distribution of HTO in the soil,

and as a consequence, on the soil characteristics (structure, texture, water content) and on the development of the root system (McFarlane et al., 1979b). Rooting patterns are dependent on the species and the development stage of the plants (McFarlane et al., 1979b). In all events, the residence time of tritium in plants growing on contaminated soil depends on the residence time of tritium in that soil (Belot, 1986). If the exposure is only from the soil and long enough, the tritium concentration in the vessels of the plants increases slowly until an equilibrium is reached with the tritium concentration in the soil water. In experiments on sunflower plants (*Helianthus annuus* L.) and tobacco plants (*Nicotiana rustica* L.) transferred to a tritiated nutrient solution, Raney and Vaadia showed that the TFWT concentration in the stem tissue and petioles of mature leaves approach equality with the tritium concentration in the nutrient solution in 12 h (Raney and Vaadia, 1965), whereas equilibrium is reached more slowly in the terminal nodes. On the contrary, the TFWT concentration in the interveinal tissue of leaves was observed to remain lower, probably because of water exchange with the atmosphere.

Moreover, it can be noted that most soil tritium (HTO) is reemitted, during vegetation period, into the atmosphere, from 1/2 to 4/5 during 12 h depending at least on light conditions. Consequently, the uptake of HTO vapour reemitted from the soil is another way of indirect contamination of the plants by the atmospheric way (Belot et al., 1988; Täschner and Bunnenberg, 1997; Yokoyama et al., 2004).

3.1.3. HTO

The absorption of H₂O (thus HTO) vapour by the aerial parts of the plants (mainly the leaves) is a diffusion phenomenon through small pores (stomata), and is controlled mostly by climatic conditions (temperature, relative humidity, light) and plant physiology (stomatal density, stomatal gating, hormonal factors) (Eisenbud et al., 1978; McFarlane et al., 1979b). A small part of the water exchange (usually less than 10%) may also occur directly by passive diffusion through the cuticle, which is the external layer limiting the aerial organs of higher plants (see below). Under adequate conditions of moisture for C3 plants (accounting for more than 95% of earth's plant species, and so most edible plants), stomata are open in the day and more or less closed at night. Consequently, the uptake of HTO from the atmosphere has been found to occur easily in the daylight (even with strong transpiration of the plants), and more slowly in the dark for various plants (Kline and Stewart, 1974; McFarlane et al., 1979b; Garland and Cox, 1982; Couchat et al., 1983; Spencer, 1984; Belot, 1986; Murphy, 1993; Diabaté and Strack, 1997). Inside the sub-stomatal cavity, HTO vapour becomes part of the solvent system by incorporating the liquid water layer of the cavity. Tritium is then subject to transpiration or transport away from stomata to the whole plant by diffusion (McFarlane et al., 1979b; Murphy, 1993;

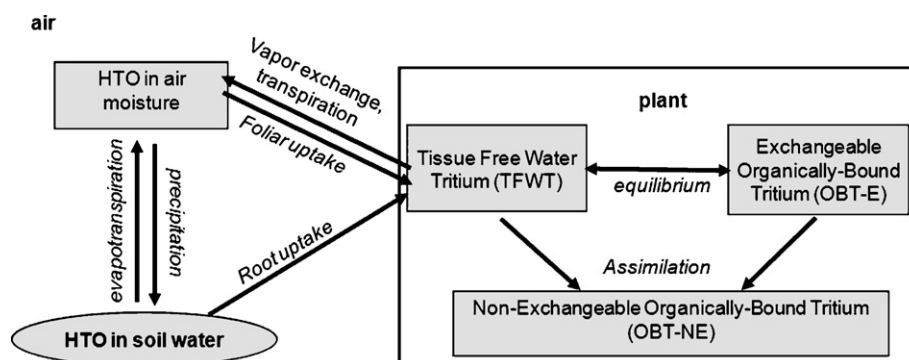


Fig. 2. General scheme of tritium transport and cycling in the vegetation.

IAEA, 2007b). According to the transpiration rate of the plants, the residence time of tritium in leaves varies (Cline, 1953; Aronoff and Choi, 1963; Raney and Vaadia, 1965; Koranda and Martin, 1971; Kline and Stewart, 1974; Eisenbud et al., 1978; McFarlane et al., 1979b; Amano and Garten, 1991; Raskob, 1995). For instance, under low conditions of vapour pressure deficit between the atmosphere and the leaves (relative humidity close to 1), the transpiration, controlled by evaporative demand, is slow. Consequently, the water (and thus the tritiated water) uptake by the rooting system from the soil to the aerial organs is limited. Then, diffusion from leaf stomata is the only way of contamination by tritium (McFarlane et al., 1979b; Diabaté and Strack, 1997; IAEA, 2007b). As the relative humidity drops, the vapour pressure deficit and the transpiration phenomena increase to a certain extent, diluting the input from the atmosphere. At the limit where the relative humidity is nil (physically impossible), there is no HTO in the air and no diffusion transfer from the air to the leaves. As a consequence, the TFWT in the foliar parts tends towards the tritium concentration in the transpiration stream, that is to say the tritium concentration in the water of the soil (IAEA, 2007b). McFarlane et al. (1979b) found that under most agricultural conditions, contamination due to rooting pattern is greater than foliar contamination. On the other hand, some authors suggested on the contrary that TFWT concentration in the plants is mainly due to atmospheric contributions (IAEA, 2007b). Raskob (1995) mentioned uptake by the atmospheric route about four times higher than the contribution of the soil (Raskob, 1995). This variability in the water uptake and exchange routes according to the water status of the plants and to the environmental conditions may explain the discrepancies in experimental data found in the literature.

However, in the real environment, part of the atmospheric tritium settles in the ground and contaminates the soil. Thus, even when the source of tritium is originally only in the atmosphere, the soil quickly becomes another source of contamination for the plants (Dinner et al., 1980; Amano and Garten, 1991). In this case, the tritium concentration in the soil water is generally lower than the atmospheric level, which leads to the dilution of HTO in the leaf water due to the transpiration stream. Therefore, plants exposed to tritiated water vapour fail to achieve an equivalent concentration of HTO in the TFWT pool, even after exposures long times (Eisenbud et al., 1978; Belot, 1986; Amano et al., 1995).

Furthermore, the tritiated water concentration in plants has been proved to be higher than the average concentration in rainfall but lower than the average concentration in atmospheric water vapour (Murphy et al., 1992). Indeed, the HTO concentration in raindrops cannot achieve its equilibrium value during precipitation, so the specific activity of tritium in rain remains lower than in the moisture of the air (Amano and Kasai, 1988; Belovodski et al., 1997). Because plants take up tritium from both atmospheric humidity and soil water resulting from precipitation, the ensuing TFWT concentration is intermediate between the tritium levels in the water vapour of the air and in the soil water (Murphy, 1993).

3.1.4. Cuticular transfer of tritium in plants

In all plants with an aerial part, a cuticle protects the epidermis and prevents the plants from the loss of water. Although such a cuticle has a very low permeability, it allows gas and vapour exchange. For instance, about 5–10% of the water loss occurs through the cuticle. Consequently, a small transfer of tritiated water vapour due to diffusion through the plasma cuticles of plants leaves seems to be theoretically possible. The transfer of tritiated water through cuticle is included in the parameter of “maximum leaf resistance/minimum conductance” in models as UFOTRI or FDMH in RODOS. UFOTRI considers a single value 10 times higher than minimum leaf resistance while experimental data shows a large variability.

3.1.5. Particular cases

3.1.5.1. C4 plants. Except in dry tropical regions, C4 plants such as maize (*Zea mays* L.), sugar cane (*Saccharum officinarum* L.) and sorghum (*Sorghum bicolor*), are very much less numerous than C3 plants. Their biological mechanisms concerning photosynthesis are quite different; these plants are able to keep a high photosynthetic level even during low water stress inducing stomatal closure and thus low CO₂ concentration. Their low transpiration rate diminishes the influx of water from the roots and the closure of the stomata prevents diffusion between tissue water and atmospheric water vapour. Consequently, for the same conditions of steady-state exposure, the TFWT concentration observed in C4 plants is a little lower than in C3 plants (Kim and Baumgärtner, 1994).

3.1.5.2. Non vascular plants and fungi. Absorption of tritiated water by plants such as mosses, lichens or fungi occurs rapidly (Eisenbud et al., 1978; Golubev et al., 2003). The surface of these plants, in contrast to higher plants, has not developed a cuticle or stomatal apparatus able to control the processes of water vapour exchange. For instance, the lichen thallus is permanently open allowing the uptake of significant quantities of water and dissolved elements from the environment (Golubev et al., 2003). Moreover, such organisms are well known to have a very high and specific metabolism, which leads to the quick absorption of surface-deposited elements such as radioactive and chemical fallout. Studies of the incorporation of tritium by lichens around nuclear facilities have shown that higher quantities can be fixed by such organisms than by green plants (Ichimasa et al., 1989; Golubev et al., 2002a; Daillant et al., 2004b; Daillant et al., 2004a). On one hand, the metabolism of these organisms is still not well understood, on another hand, important decrease of the atmospheric source occurred during the last 30 years, as a consequence, any conclusions as to environment contamination with regard to tritium levels in lichens remains speculative (Vichot et al., 2008).

3.1.5.3. Other cases. Some plant types with particular ecophysiological and morphological features (tall forbs, heliophytes, reeds...) display high transpiration rates ranged between 3 and 10 mmol H₂O m⁻² s⁻¹ (Larcher, 2003). Consequently, they are able to exchange large amounts of water in a very short time. Although no experimental data are currently available on HTO uptake in such species, they may also display unusual behaviour concerning tritium exchange.

3.1.6. Kinetics of tritiated water uptake from the environment to the plants

Due to the difficulty of making long-term experiments with tritium, studies of chronic exposure have mainly been carried out in real conditions on the vegetation around nuclear facilities (Murphy et al., 1982; Amano and Kasai, 1988; Amano and Garten, 1991; Murphy et al., 1992; Amano et al., 1995; Evenden et al., 1998; Hisamatsu et al., 1998; Davis et al., 2002; Davis et al., 2005b). These studies have led to the conclusion that tritium is present in all water pools (in plants, animals, soil, etc.) and will tend to equilibrium, when the duration of tritium exposure is stable and sufficiently long.

Tritium concentrations in plants result from fast equilibration between the different plant compartments (rooting system and aerial parts), and environmental compartments (atmosphere and soil). Numerous studies have been carried out into the kinetics of tritium uptake by plants, the majority focusing on the transfer from atmospheric HTO to plants. They have defined several ways to express the uptake or loss rate and the steady-state kinetics.

Different models and equations have been proposed to express the uptake kinetics of tritiated water. Experimental data in various plants such as tomato (*Solanum lycopersicum* L.), sunflower (*H.*

Table 2

Characteristic times required to reach equilibrium between the tissue free water tritium (TFWT) concentration in plants and the HTO vapour concentration in the atmosphere (t) or to reach one-half of the TFWT steady-state concentration ($t_{1/2}$), uptake rate constants (k) and TFWT/atmospheric HTO ratios for atmospheric exposure to HTO on various plants, under steady-state and daylight conditions.

Reference	Plant	Plant part	$t_{eq1/2}$ (h)	t_{eq} (h)	k (h^{-1})	TFWT/HTO ratio
Murphy (1993)	Vegetation	All	–	0.25–2	–	0.77–0.82
Choi et al. (2002)	Rice (<i>Oryza sativa</i> L.)	All	–	1	–	–
Guenot and Belot (1984)	Potato plants (<i>Solanum tuberosum</i> L.) wine grape plants (<i>Vitis vinifera</i> L.)	Leaves	–	0.75	–	–
Kline and Stewart (1974)	Grass	Leaves	1.1	–	0.63	–
		Stems	33	–	0.02	–
Spencer (1984)	Tomato plants (<i>Solanum lycopersicum</i> L.)	New foliage	29–37	–	0.019–0.024	0.43
		Old foliage	7	–	0.104	0.46
		Green fruits	17–69	–	0.010–0.042	0.19
Davis et al. (2002)	Lettuce plants (<i>Lactuca sativa</i> L.)	Leaves	–	–	–	0.5
	Tomato plants (<i>Solanum lycopersicum</i> L.)	Fruits	–	–	–	0.68
	Grass	All	–	–	–	0.32–0.84
Dinner et al. (1980)	Tomato plants (<i>Solanum lycopersicum</i> L.) cucumber plants (<i>Cucumis sativus</i> L.)	Fruits	–	–	–	0.4–0.8

annuus L.), or grass were found to follow an exponential first-order law (2) during atmospheric exposure (Kline and Stewart, 1974; Couchat et al., 1983; Spencer, 1984).

$$C_{TFWT} = C_{\infty}(1 - e^{-kt}) \quad (2)$$

where C_{TFWT}^t : TFWT concentration in the plant at the considered time t ($Bq L^{-1}$); C_{∞} : steady-state TFWT concentration ($Bq L^{-1}$); k : rate constant for HTO uptake (h^{-1}); t : time after the beginning of exposure (h).

Here, the time required to reach equilibrium between the TFWT concentration in plants and the atmospheric concentration (t_{eq}) is usually given to quantify the tritium uptake. It is important to note that the term “equilibrium” does not correspond to the complete equilibration between the TFWT and the atmosphere activities, but to a steady-state. Indeed, as explained previously, the TFWT concentrations always remain lower than the atmospheric HTO activities, even under constant long-term exposure conditions (Eisenbud et al., 1978; Spencer, 1984). When the tritium source is located only in the atmosphere, the equilibrium is reached within a few hours for stable and daylight environmental conditions. The time necessary to reach one-half of the steady-state concentration ($t_{eq1/2}$) is also used (Table 2).

The uptake rate constant (k) is another way to present tritium absorption ($k = 0.693/t_{eq1/2}$). This rate is typically lower than $1 h^{-1}$. However, the uptake rate is dependent on the time of the day when the tritium exposure occurs; as mentioned previously, the absorption of atmospheric HTO via the stomata is much faster in daylight than in the night (Couchat et al., 1983).

Another way to quantify the uptake of tritium into plants at equilibrium is to calculate the average ratio of TFWT concentration to HTO concentration in the air for steady-state conditions. As the equilibrium is never reached, this ratio is always lower than 1. Moreover, Davis et al. (2002) noticed on grass samples a strong diurnal variation of this uptake ratio, due to the variation of relative humidity in the daytime (Davis et al., 2002).

Some experimental data given in the literature for characteristic times, tritium uptake rates and ratios of TFWT to atmospheric HTO, under steady-state and daylight conditions, are gathered in Table 2 (Kline and Stewart, 1974; Dinner et al., 1980; Guenot and Belot, 1984; Spencer, 1984; Murphy, 1993; Choi et al., 2002; Davis et al., 2002).

Irrespective of how the calculation is done, it should be noticed that the tritium uptake rate depends on the plant species, the plant organ, and the biological stage of development during atmospheric exposure. For instance, Choi et al. (2002) found higher HTO concentrations at equilibrium in the leaves and the ears of rice (*Oryza*

sativa L.), and lower in stem, varying with plant parts by a factor of up to 10. Amano et al. (1995) reported higher HTO concentrations in petioles and leaflets of tomato than in fruits. According to the TRS-364 (IAEA, 2007b), the plant leaves draw the majority of their tritium (as TFWT) from the air in normal circumstances. In contrast, the fruits, tubers or root crops draw a large fraction of their water from the soil, and so the tritium-free water concentration in these organs is generally lower than in leaves. However, the data available do not enable an evaluation of the tritium concentration in the water of these organs with any confidence (IAEA, 2007b).

3.2. Kinetics of tritium loss after its uptake by plants

After atmospheric exposure to tritium, the residence time of TFWT in plants is linked to the renewal of water in the plant, and is generally short (Elwood, 1971). The rate of TFWT concentration decrease is usually described as the sum of three exponentials (Belot, 1986; Diabaté and Strack, 1990; Brudenell et al., 1997); given in Eq. (3).

$$C_{TFWT}^t = C_{TFWT}^0 \times [1 - \exp(-k_1 t) - \exp(-k_2 t) - \exp(-k_3 t)] \quad (3)$$

where C_{TFWT}^t : TFWT concentration in the plant at the considered time t ($Bq L^{-1}$); C_{TFWT}^0 : initial TFWT concentration in the plant immediately after exposure ($Bq L^{-1}$); k_1, k_2, k_3 : characteristic rate constants of tritium release corresponding to the decrease of the TFWT concentration described as a sum of three exponentials (h^{-1}).

In such models based on experimental data, the first component accounts for more than 99% of the initial concentration of the TFWT compartment. This fraction is released very quickly with a short half-life T_1 of less than one hour (McFarlane et al., 1979b; Belot, 1986; Brudenell et al., 1997), due to the transpiration of the plant. The second component of exponential models is related to less accessible HTO (colloidally bound water), to exchangeable OB (Cline, 1953; Choi et al., 2002) or to decontamination of stem and root tissues labelled by phloem translocation (Guenot and Belot, 1984; Brudenell et al., 1997). It accounts for less than 1% of the absorbed tritium (Belot, 1986) and has a half-life T_2 of several tens of hours. A third and long-lived component could be observed and corresponds to non-exchangeable OB. Although it stands for a tiny part of primarily absorbed tritium, the decrease is very slow with a half-life T_3 of several hundreds of hours. Considering that the first component is predominant, the characteristic rate constant of tritium release (k_1) is usually calculated according to a single exponential law. The half-lives of tritium and the release rate constant associated for such exponential models were determined for various plants (Koranda and Martin, 1971; Anspaugh et al., 1973; Kline

Table 3
Residence half-times (T_1 , T_2 , T_3) for tritium in plants according to an exponential model with one to three compartments, and characteristic constant rates of tritium release corresponding to the first compartment (k_1). Data were determined experimentally for daylight conditions.

Reference	Plant species	T_1 (min)	k_1 (min^{-1})	T_2 (h)	T_3 (h)
Guenot and Belot (1984)	Potato plants (<i>Solanum tuberosum</i> L.) wine grape plants (<i>Vitis vinifera</i> L.)	30	0.023	30	Hundreds
Brudenell et al. (1997)	Cabbage plants (<i>Brassica oleracea</i> L.)	53	0.013	15	48
	Lettuce plants (<i>Lactuca sativa</i> L.)	53	0.013	6	–
Anspaugh et al. (1973)	–	45	0.015	25	–
Koranda and Martin (1971)	Alfalfa (<i>Medicago sativa</i> L.)	20	0.035	1	270
Kline and Stewart (1974)	Grass vegetation (stems)	25	0.028	–	–
	Grass vegetation (leaves)	35	0.020	–	–
Keum et al. (2006)	Ear of rice (<i>Oryza sativa</i> L.)	30	0.023	–	–
Iyengar et al. (1981)	Lettuce plants (<i>Lactuca sativa</i> L.)	32	0.022	–	–
	Cabbage plants (<i>Brassica oleracea</i> L.)	45	0.0154	–	–
	Shoots of hot pepper (<i>Capsicum annuum</i> L.)	46	0.0152	–	–
	Roots of hot pepper (<i>Capsicum annuum</i> L.)	65	0.0107	–	–

and Stewart, 1974; Iyengar et al., 1981; Guenot and Belot, 1984; Brudenell et al., 1997; Keum et al., 2006); some examples are given in Tables 3 and 4.

Tritium delabeling of plant tissue after exposure from the atmosphere is much slower in darkness than in the light, loss of HTO primarily depending on transpiration (Kline and Stewart, 1974; Brudenell et al., 1997). Moreover, the discrepancy in the rate of the TFWT loss observed among plant species is probably due to physiological and morphological differences. Moreover, the environmental conditions at the time of the measurements could also explain the discrepancies.

3.3. Incorporation of tritium into plants organic matter

The different chemical forms of tritium are not incorporated as OBT in the same way. Above all, HTO is most relevant to the incorporation of OBT in green plants due to photosynthesis (Belot, 1986; Diabaté and Strack, 1993). As described previously, the majority of the tritium absorbed by the plants returns to the atmosphere because of transpiration. The small remaining fraction is available for biological processes, especially photosynthesis. Since the fraction of hydrogen converted into organic matter ranges from 0.06% to 0.3% for growing crops (Kramer, 1969; Murphy, 1990), the amount of tritium incorporated into organic matter is small compared to that moving through the plant as tritiated water.

3.3.1. Mechanisms of OBT production in plants

The first studies about tritium incorporation into organic matter were carried out by Moses and Calvin in 1959 (Moses and Calvin, 1959). Their work dealt with photosynthesis in single-cell algae, *Chlorella pyrenoidosa*. After a short exposure to HTO and $^{14}\text{CO}_2$, tritium was identified in chemical species corresponding to the first photosynthetic products (hexoses, carbohydrate monophosphates and diphosphates, organic acids and amino acids). They proved that tritium could be integrated in the dark into molecules of the tricarboxylic acid cycle and derived amino acids, but at low rates. Moreover, the authors noticed that the molecules labelled with tritium were also marked with ^{14}C upon simultaneous exposure

to the two radionuclides. Since the 1950s, numerous studies concerning the use of $^3\text{H}_2\text{O}$ and $^{14}\text{CO}_2$ as radioactive tracers to learn about the photosynthetic processes have been published (Biddulph and Cory, 1957; Gage and Aronoff, 1960; Aronoff and Choi, 1963; Choi and Aronoff, 1966; Trip and Gorham, 1968a, b; Thompson and Nelson, 1971; Peterson, 1982; Guenot and Belot, 1984) and have confirmed that photosynthesis is the most important process in the assimilation of non-exchangeable OBT.

3.3.1.1. OBT production from HTO. Diabaté and Strack (1993) have detailed the biological processes leading to OBT formation. Various and complex biochemical reactions occur during the steps of photosynthesis. Some reactions need light energy to occur: they are called “light reactions”. The reactions that are not of a photochemical nature are called “light-independent reactions”. During the daylight, the photolysis of water and of HTO if present produces reduction equivalents $\text{NADPH} + \text{H}^+$, and possibly T^+ and energy equivalents (ATP) inter alia. These compounds can then be used in the light-independent reactions of the Calvin cycle and lead to the production of phosphoglycerate: this is the primary reaction in which hydrogen or tritium is integrated into an organic molecule. After this first step of carbon fixation, phosphoglycerate reduction generates glyceraldehyde-3-phosphate (G3P), which is partially exported out of the chloroplast to form glucose and other organic compounds in the cytosol of the cell (Moses and Calvin, 1959). Protons H^+ (or T^+) could then be incorporated into various chemical bonds such as $-\text{C}-\text{H}$ or $-\text{C}-\text{T}$, $-\text{S}-\text{H}$ or $-\text{S}-\text{T}$, $-\text{N}-\text{H}$ or $-\text{N}-\text{T}$, $-\text{P}-\text{H}$ or $-\text{P}-\text{T}$.

Other biochemical pathways such as those of oxidative respiration or the tricarboxylic acid cycle lead to the incorporation of tritium into OBT regardless of the lighting level (Moses and Calvin, 1959; Guenot and Belot, 1984; Belot, 1986; IAEA, 2007b). These “dark reactions” use the substances built up by light reactions; they correspond to the use of organic molecules for the needs of the plant (energy, growth, maintenance) and not to autotrophic synthesis (Diabaté and Strack, 1997). On the other hand, a multitude of reactions of hydrogenations and dehydrogenations occur at various stages of the plant metabolism. They lead to the transfer of

Table 4
Residence half-times (T_1 , T_2 , T_3) for tritium in plants according to an exponential model with one to three compartments, and characteristic constant rate of tritium release corresponding to the first compartment (k_1). Data were determined experimentally for darkness conditions.

Reference	Plant	T_1 (min)	k_1 (min^{-1})	T_2 (h)	T_3 (h)
Kline and Stewart (1974)	Grass vegetation (stems)	400	0.0017	–	–
	Grass vegetation (leaves)	2180	0.0003	–	–
Brudenell et al. (1997)	Lettuce plants (<i>Lactuca sativa</i> L.)	780	0.0009	–	–

hydrogen atoms (or tritium) from one molecule to another. OBT formation in these ways is much lower than with the light reactions but significant. At night, the rate of OBT formation drops to 1/5 to 1/3 of the daytime rate (Thompson and Nelson, 1971; Diabaté and Strack, 1993, 1997; Galeriu et al., 2008). However, OBT formation during the night time is still not very well understood and will very likely be influenced by external factors as higher air concentrations, intensity of stomatal opening and so on. This point has to be investigated in the future.

3.3.1.2. OBT production from HT. Tritium in the gas form HT may be involved in processes other than photosynthesis leading to the production of organic matter (Elwood, 1971; McFarlane, 1978; Sweet and Murphy, 1984; Belot, 1986). However, the deposition velocities of HT into OBT are very low (0.1×10^{-11} to 4.6×10^{-11} m s⁻¹ were found for lettuce, tomato, cabbage, maize and grass leaves Strack et al., 1991), which led to this route of OBT synthesis being ignored in predictive tritium dose models (Diabaté and Strack, 1993).

As described previously, the most common way for plants to become contaminated by HT involves its oxidation into HTO in plant foliage or in the soil (Stewart, 1971; McFarlane et al., 1978, 1979a; Belot, 1986; Bunnenberg et al., 1986; Spencer and Dunstall, 1986; Diabaté and Honig, 1988; Foerstel, 1988; Amano et al., 1995; Ichimasa et al., 1999).

3.3.1.3. OBT production from other tritiated compounds. Tritiated methane may also be directly involved in the synthesis of OBT by vegetation (Amano, 1995). However, the exposure to tritiated methane is quantitatively very low (Mason et al., 1971; McFarlane et al., 1979b; Diabaté and Strack, 1993).

Some authors reported that tritiated formaldehyde is quite easily incorporated to the non-exchangeable fraction of OBT due to its high solubility in plant tissue water and high affinity to plant organic matter (Diabaté and Strack, 1993). In plants, formaldehyde is bound to proteins in the cell membranes and to the free amino acids inside the cell. However, the percentage of OBT formed in this way is very small because formaldehyde concentrations are naturally low and human release of tritiated formaldehyde is negligible.

3.3.2. Translocation of OBT in plants

Many mechanisms and molecules are implied in carbon allocation. In a preliminary simplified approach, we can consider that organic molecules built up during the photosynthetic light reactions are either stored as starch in the chloroplasts, or exported to the cytosol where they are converted into sucrose, mainly during the night (Diabaté and Strack, 1993). In the phloem, the photoassimilates (in the form of sucrose) are transported from the leaves to the whole plant according to the needs for growth, energy storage, fruit production, etc. This phenomenon is usually called “translocation” (Thompson and Nelson, 1971; Belot, 1986). Sucrose is then split into glucose and fructose, which are directly available for energy production and as structural or storage compounds. As an isotope of hydrogen, tritium is integrated into such molecules to form OBT. Its incorporation and translocation rates then depend on the development stage of the plant at the time of exposure (Diabaté and Strack, 1992, 1993). The organs concerned and rates of the translocation of OBT vary with seasonal changes. For example, in the vegetative step, translocation to fruits or seeds is low, whereas the photoassimilated fluxes are higher during fruit growth (Diabaté and Strack, 1993). Indeed, the organic matter of fruits and tubers is totally imported from photosynthesis sites.

To quantify translocation, Diabaté and Strack (1997) defined a translocation index (TLI) in wheat. The TLI represents the percentage of the OBT concentration in grain water combustion at harvest related to the TFWT concentration in leaves at the end of exposure to HTO. The experiments confirmed that TLI varies according

to plant development at the time of exposure, from 0.06% to 0.2% for HTO exposure on very young plants to 0.4–0.9% for HTO exposure on plants in a linear phase of seed growth. TLI then decreases to 0.04–0.2% when maturity begins (Diabaté and Strack, 1997).

3.3.3. Factors controlling OBT production in plants

Like any biological phenomenon, the production of OBT depends on the plant's stage of development at the time of exposure. Consequently, the amount of OBT incorporated depends on the plant species, the part of the plant considered, and on the biological stage during which the exposure takes place (Indeka, 1981; Diabaté and Strack, 1993; Amano et al., 1995).

3.3.3.1. Plant species and plant organs. The particular biological characteristics of each plant species and the specific cultivation techniques lead to differences in OBT incorporation. Indeed, OBT is formed by photosynthesis in a small fraction of the TFWT leaf compartment. The assimilates produced are first used for the growth and development of the leaves, roots and stems. Finally, the organic molecules generated can be used or stored in the fruits and the tubers. The translocation leads to the dispersion of organic molecules throughout the whole plant and to discrepancies in OBT concentration according to the plant species and to the part of the plant considered (Spencer, 1984; Diabaté and Strack, 1997). In particular, the fruits and tubers accumulate a lot of carbon molecules: they act as OBT sinks. For instance, Atarashi-Andoh et al. (2002) studied the conversion rate, defined as the proportionality coefficient between OBT production and TFWT concentration, for several vegetables: komatsuna (*Brassica rapa* var. *pervidis* or var. *komatsuna*, commonly named Chinese mustard), radish (*Raphanus sativus* L.) and cherry tomato (*S. lycopersicum* L.). They calculated the conversion rate from HTO in plant leaves to OBT in each of the plant organs and found that the conversion rates in fruits and roots were smaller than in leaves, because OBT was produced in leaves and then translocated to fruits and roots.

3.3.3.2. Biomolecules. The rate of OBT formation depends on the metabolic activity and the turnover rate of the biomolecules which contain tritium (Diabaté and Strack, 1993). As all organic matter in green plants is produced by photosynthetic mechanisms, tritium is incorporated as OBT into complex molecules like polysaccharides, proteins, lipids and nucleic acids (Diabaté and Strack, 1993). While in proteins, the theoretical hydrogen content is about 6.8%, with 75% in non-exchangeable positions, this fraction is estimated to be about 12% for lipids, with 95–100% being non-exchangeable (Diabaté and Strack, 1993). The variations in quantitative and qualitative biochemical composition from one species to another leads to large differences in the formation of exchangeable and non-exchangeable hydrogen, and therefore in OBT production.

3.3.3.3. Developmental stage. Several authors have shown that OBT incorporation is dependent on the growth and development stage of the plant (Choi et al., 2000; Atarashi-Andoh et al., 2002; Choi et al., 2002; Choi et al., 2005; Vichot et al., 2008; Boyer et al., 2009). For instance, Amano et al. (1995) reported that young leaves of komatsuna (Chinese mustard) showed higher OBT concentration than old leaves after a 12-day exposure to HT. The lower content of water and the higher photosynthetic activity of young leaves explain the higher OBT production. In another experiment (Boyer et al., 2009), the foliar uptake of tritium and its incorporation as OBT in lettuces continuously exposed to atmospheric HT and HTO was studied from the earliest growth stages to plant senescence. The conversion rate from TFWT to OBT in plant leaves was calculated from weekly measurements. It was observed to vary significantly during the lettuce growth, with a significant increase in the stages with the highest growth rates.

3.3.3.4. *Light and CO₂*. Because the photosynthesis is modulated by environmental factors (light, temperature, humidity, CO₂ and O₂ levels) and plant features (species, physiological state, hormonal factors), the transfer of tritium from TFWT to organic matter and the production rate of OBH is quantitatively difficult to predict (Diabaté and Strack, 1993; Atarashi-Andoh et al., 2002). For Diabaté and Strack (1993), a reasonable approach to assess the production of OBH is based on the measurement of tritium concentration in the air and the estimation of the photosynthetic rate via the measurement of CO₂ exchange between the atmosphere and the plants. Photosynthetically active radiation (PAR), usually expressed as photosynthetic photon flux density ($\mu\text{mol m}^{-2} \text{s}^{-1}$) could also be taken into account to assess the photosynthetic activity of the plant and subsequently the OBH production (Diabaté and Strack, 1997; Atarashi-Andoh et al., 2002).

3.3.4. Kinetics of OBH production and turnover

Whereas TFWT becomes equilibrated very quickly with environmental HTO, tritium incorporated into organic matter increases slowly. The specific ratio of OBH to OBH in plants increased gradually from the start of exposure but this organic compartment does not usually reach an equilibrium with the tritium content of tissue water even following long-term exposure (Diabaté and Strack, 1993). For instance, the experiment reported by Amano et al. (1995), who exposed different plant species (cherry tomato, radish, komatsuna) to HT for 12 days, was not long enough to attain a steady state for OBH.

The turnover of OBH, particularly in storage organs, is slow (Diabaté and Strack, 1993, 1997). Following a short burst of exposure, the total amount of tritium assimilated as OBH decreases slowly throughout the life of the plant. In contrast, the specific activity declines significantly due to the production of new organic matter and then dilution of OBH in plant material.

3.3.5. Tritium fractionation in OBH

Although tritium concentration due to isotope discrimination has been proved to be negligible on the ecosystem scale (Elwood, 1971; Rohwer and Wilcox, 1976), the difference in the atomic weight of protium and tritium leads to significant isotopic effects in OBH synthesis and catabolism (Belot, 1986; Diabaté and Strack,

1993). Chemical reactions occur at lower rates when tritium is involved than reactions involving protium. As a consequence, less tritium is incorporated into organic molecules than hydrogen but, on the other hand, tritium fixed to carbon is cleft more slowly than C–H bonds (Diabaté and Strack, 1993). Detailed literature is available on the mechanisms of organic tritium fractionation in plants (McFarlane, 1976; Kim and Baumgärtner, 1991, 1994). When potato plants (*Solanum tuberosum* L.) and wine grape plants (*Vitis vinifera* L.) were exposed to tritium, Guenot and Belot (1984) showed that the tritium incorporation into OBH was 30% smaller than predicted according to the photosynthetic rates. Davis et al. found a significant isotopic discrimination factor in OBH formation from the associated TFWT fraction for different plants: 0.52 for grass, 0.68 for lettuce, 0.46 for radish roots and 0.72 for tomato fruits (Davis et al., 2002).

The specific activity ratio (SAR) is the ratio of OBH to HTO in a given compartment; it is an indicator of the ability of biological systems to concentrate tritium into the organic fraction (Okada and Momoshima, 1993). As the ratio is formulated in terms of tritium concentrations in water, OBH concentration corresponds to the activity in the water produced by complete combustion of the dry material, called “water equivalent”. Generally, tritium incorporation is compared to that of protium by the ratio of the specific tritium activity of hydrogen in OBH of an organic compound to the HTO activity in the environment of the considered organisms (Diabaté and Strack, 1993). The OBH/TFWT ratio could also be used. According to these definitions, some authors report values for OBH/HTO in air moisture and OBH/TFWT ratios for various plants for long-time exposure to atmospheric tritium (McFarlane, 1976; Garland and Ameen, 1979; Spencer, 1984; Hisamatsu et al., 1987; Hisamatsu et al., 1989; Takashima et al., 1989; Inoue and Iwakura, 1990; Davis et al., 2002; Davis et al., 2005b). A non-exhaustive list is given in Table 5. The high variability of these ratios is explained by very different experimental conditions. Under steady-state conditions, the values lower than 1 obtained in most cases are probably related to the discriminatory isotopic effect against tritium during the formation of organic matter (Spencer, 1984). However, equilibrium conditions are generally not reached during plant growth or at the time of sampling. Because of the longer residence time in plants of OBH compared to TFWT, the specific activity ratios observed could

Table 5

Specific activity ratios between organically bound tritium (OBH) concentration and tritium air moisture concentration (HTO), and between OBH and tissue free water tritium concentration (TFWT), for various plants.

Reference	Plant	OBH/HTO ratio	OBH/TFWT ratio
McFarlane (1976)	Alfalfa (<i>Medicago sativa</i> L.)	0.78	–
Garland and Ameen (1979)	Barley (<i>Hordeum vulgare</i> L.)	0.73	–
	Maize (<i>Zea mays</i> L.)	0.64	–
Davis et al. (2002, 2005b)	Grass	0.19–0.31	0.52
	Lettuce leaves (<i>Lactuca sativa</i> L.)	0.41	0.68
	Radish roots (<i>Raphanus sativus</i> L.)	0.25	0.46
	Tomato fruits (<i>Solanum lycopersicum</i> L.)	0.43	0.72
	Forage crops	0.33–0.56	–
	Maize (<i>Zea mays</i> L.)	0.14–0.39	–
	Barley (<i>Hordeum vulgare</i> L.)	0.52–0.57	–
	Spring wheat (<i>Triticum aestivum</i> L.)	0.18–0.25	–
	Fruit vegetables	0.32–0.37	–
Takashima et al. (1989)	Pine needles (<i>Pinus thunbergii</i> and <i>Picea jezoensis</i>)	–	0.9–2.1
Hisamatsu et al. (1987, 1989)	Fruits	–	1.3–2.4
	Green vegetables	–	1.0–1.5
	Polished rice (<i>Oryza sativa</i> L.)	–	0.57–1.3
Inoue and Iwakura (1990)	Rice (<i>Oryza sativa</i> L.)	–	0.83
Spencer (1984)	Tomato plants: new foliage (<i>Solanum lycopersicum</i> L.)	0.30	0.70
	Tomato plants: old foliage (<i>Solanum lycopersicum</i> L.)	0.29	0.63
	Tomato plants: green fruits (<i>Solanum lycopersicum</i> L.)	0.27	0.72–1.92

be higher than 1 (Inoue and Iwakura, 1989). Moreover, the ratio of OBT to TFWT is usually higher than the ratio of OBT to atmospheric HTO, because the TFWT concentration never reaches equilibrium with the HTO concentration in air moisture.

The particular biological mechanisms of each plant species and organs could also explain the variability in OBT/HTO and OBT/TFWT ratios. Fruits and the tubers store the organic assimilates (i.e. OBT), whereas this is not the main function of leaves or roots (Diabaté and Strack, 1993). Consequently, the values higher than 1 found for tritium-specific activity ratios in some organs like fruits (Spencer, 1984; Hisamatsu et al., 1987; Hisamatsu et al., 1989) may not be attributed to tritium enrichment but to the translocation of foliar OBT to growing organs.

Moreover, particular cultivation techniques could explain the low ratios found in some plant species, like rice. In this case, flooding leads to equilibrium between the tissue water of the plant and the irrigation water. The fluctuations of tritium concentration in the atmosphere are then negligible (Inoue and Iwakura, 1990; Diabaté and Strack, 1993), this explains the very low ratio of OBT to atmospheric HTO.

3.3.6. Transfer functions

Tritium transfer from the environmental HTO or the TFWT pool to the OBT pool have been calculated using various approaches.

Choi et al. (2000) used the following definition for tritium exposure of rice by watering (4):

$$TF = \frac{C_{OBT}}{C_{HTO}} \quad (4)$$

where C_{OBT} : plant dry matter concentration at harvest ($Bq\ kg^{-1}$); C_{HTO} : decay-corrected HTO activity applied to the harvest ($Bq\ m^{-2}$).

Atarashi-Andoh et al. (2002) have studied the OBT formation in the case of a continuous HT release. In this case, the source of plant HTO is mainly soil. After few days from start experiment, there is a quasi-equilibrium in soil water HTO (except rain events) and daily averaged plant HTO is not very variable. Under these circumstances, they proposed a definition of a conversion rate, following (5):

$$\frac{dC_{OBT}}{dt} = \nu C_{TFWT}(t) \quad (5)$$

where C_{TFWT} : concentration in the TFWT pool ($Bq\ L^{-1}$); C_{OBT} : steady-state OBT concentration ($Bq\ L^{-1}$); ν : conversion rate from HTO in leaves to OBT ($\%h^{-1}$)

Using this definition, the conversion rate from the TFWT to the OBT pool was determined experimentally for different plants, and the results are gathered in Table 6 (Atarashi-Andoh et al., 2002; Vichot et al., 2008; Boyer et al., 2009). The conversion rates given in Table 6 were calculated from the beginning of exposure to plant

Table 6
Conversion rates between tissue free water tritium and organically bound tritium calculated for different vegetables under steady-state conditions of exposure.

Reference	Plant	Conversion rate ($\%h^{-1}$)
Atarashi-Andoh et al. (2002)	Komatsuna leaves (<i>Brassica rapa</i> var. komatsuna)	0.20–0.32
	Radish leaves (<i>Raphanus sativus</i> L.)	0.21
	Radish root (<i>Raphanus sativus</i> L.)	0.07
	Cherry tomato leaves (<i>Solanum lycopersicum</i> L.)	0.13–0.19
	Cherry tomato fruit (<i>Solanum lycopersicum</i> L.)	0.05
Vichot et al. (2008)	Lettuce leaves (<i>Lactuca sativa</i> L.)	0.20–0.24
Boyer et al. (2009)	Lettuce leaves (<i>Lactuca sativa</i> L.)	0.16

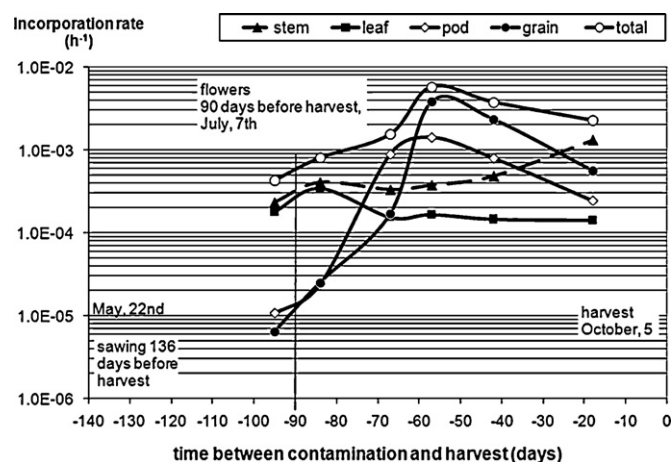


Fig. 3. OBT incorporation in different organs of soybean plants, according to the developmental stage when the tritium exposure takes place and to the elapsed time between exposure and harvest.

harvest, disregarding biological parameters such as plant mass. However, the authors have proved that the conversion rate, and thus the OBT incorporation, primarily depends on the developmental stage of the plant conditions (influence of temperature, solar radiation, leaf area index, water vapour deficit, soil water deficit) and on the environmental conditions in the vegetation period (Atarashi-Andoh et al., 2002; Vichot et al., 2008; Boyer et al., 2009).

Another experiment reported by Lee for an EMRAS exercise (soybean scenario¹) dealt with soybean plants (*Glycine max*) exposed to elevated levels of airborne tritium in a glove box. The exposure was carried out acutely for one hour at various stages in the growth of the soybeans. Following exposure, the plants were removed from the glove box and cultivated as usual outdoors. The tritium behaviour in the plant body and pods was observed by sampling the various plant parts and determining the concentrations in them. The incorporation rate of HTO into OBT at different stages between flower and harvest is shown in Fig. 3 (Guétat et al., 2008a, b). Variations by a factor of 3 orders of magnitude are observed according to the organ and the developmental stage.

3.3.7. Dose impact due to ingestion of OBT

Twenty years ago, the dose impact of OBT was recognized to be significant because of its longer residence time than HTO and its uptake into the human body by direct ingestion (ICRP, 1993; Amano et al., 1995), even if the long-term OBT pool represents a very small fraction of the fresh plant material (Diabaté and Strack, 1993; Murphy, 1993). The effective dose coefficient for adults following OBT ingestion has been evaluated to 4.1×10^{-11} Sv Bq⁻¹ whereas 1.8×10^{-11} Sv Bq⁻¹ is usually considered for HTO (ICRP, 1993). These coefficients are higher for babies (3 months), respectively 1.1×10^{-10} Sv Bq⁻¹ for OBT and 6.4×10^{-11} Sv Bq⁻¹ for HTO.

In the digestion process non-exchangeable fraction is changed. Human intake is mostly in digestible form (>90%). For a European diet, 30–50% of ingested non-exchangeable OBT is converted to body HTO. Doses arising from ingested OBT are presently discussed, because the biological half times for humans could be different than those assessed by ICRP (Melintescu et al., 2007; Galeriu et al., 2008). Recent work estimates that OBT contribution to dose is 20–30% for routine emission and about 60% for accidental ones (Guétat et al., 2008a, b). However, uncertainties about mechanisms of human digestion in regards to the high variability in biological molecules ingested remains.

¹ <http://www-ns.iaea.org/projects/emras/emras-tritium-wg.htm>.

3.4. Conclusions about tritium dose impact due to the plants in the human diet

Dealing with the impact of tritium due to plants in the human diet, it is important to bear some key points in mind. First, the chemical species incorporating the tritium have to be considered. The TFWT fraction of edible plants has a short residence time, a few days, and a low dose coefficient for an adult consumer. However, the tissue-free water fraction can account for more than 95% of the fresh mass of plants; thus, the potential dose impact of tritiated water circulating in plants is not negligible. On the other hand, the OBT fraction presents a longer residence time in plants of several weeks or months, and a dose coefficient 2.5 times higher than TFWT upon human ingestion (ICRP, 1993). Nevertheless, the quantity of non-exchangeable OBT incorporated in plants is very small, corresponding to less than 1% of the tritium initially taken up as HTO. After a single exposure, much less tritium will be incorporated into the OBT fraction than into the TFWT fraction, but the relative contribution to the radiation dose in the long range could be nearly the same due to the longer half-life of OBT (Diabaté and Strack, 1993). On chronic exposure to tritium, the formation of OBT is continuous and the contribution of OBT to the dose received becomes greater with time.

Although the kinetics laws and the rate of tritium uptake or loss as well as the incorporation into the plants are well documented for short- or long-term exposure, it is not easy to evaluate the corresponding doses for human consumers. Indeed, as time goes, lots of environmental parameters (especially transfers) have to be taken into account and make the dose estimation very complicated.

Moreover, as explained previously, tritium uptake and incorporation is highly variable with the species, the organ, the stage of development, the physiological status of the plant and the environmental conditions. As a consequence, the potential health hazards vary with those same parameters. *A fortiori*, for risk assessment these variations have to be considered in terms of dose. Note that no general conclusion should be drawn from experimental data sampled for a particular plant species in a specific environment.

4. Models

To guarantee safety around tritium facilities, the consequences of possible releases into the environment should be assessed. Modeling tritium transfer in the environment and its assimilation into biological organisms is a useful way to assess the risks, especially during emergencies.

Since the 1960s, a multitude of predictive models have been proposed to assess the tritium transfer between the different compartments of the environment. Indeed, the complexity of the phenomena involved implies specific models for each of the different steps of tritium cycling. It is not in the scope of this review to describe all models dealing with atmospheric dispersion, tritium deposition, or migration into the soil. The present overview focuses on the models most commonly used for tritium uptake and incorporation in plants, especially in agricultural vegetables. Because of the sizeable literature on this topic, a choice of the more relevant papers has been made and bibliographical references are given for further reading.

The specific activity models (SA models) hypothesize that the ratio $^3\text{H}/^1\text{H}$ remains constant from one environmental compartment to another one (IAEA, 2007b). They are useful to predict the dose engaged following chronic exposure (Anspaugh et al., 1973; Murphy, 1986; Diabaté and Strack, 1990; Hamby and Bauer, 1994; Peterson and Davis, 2002). Such models are accurate when the meteorological data and the diet of the population during exposure are known. For short-term releases, dynamic models should

be used. Some complex models such UFOTRI (Raskob, 1990, 1993; Galeriu et al., 1995; Täschner and Bunnenberg, 1997) and ETMOD (Russell and Ogram, 1992) describe the detailed fluxes of tritium through all the environmental compartments (atmosphere, soil, plants) on the condition that the meteorological conditions are known. Nowadays, it may be useful to find a compromise between complexity of the model and uncertainties and numbers of input data. As tritium is a life element, many processes and input data can and have to be taken from other life sciences researches as simple compartmental model may not be efficient enough (Keum et al., 2006), when considering the natural variability of situations within a day and through the year.

All these models consider at least three basic environmental compartments: air, soil and plants (Golubev et al., 2002b; Keum et al., 2006). In some models, these compartments are subdivided into more detailed units. For instance, plants can be classified into different categories such as leafy vegetables, root vegetables, seed, tuber plants, fruits, etc. (Raskob, 1993; Keum et al., 2006) The soil can be divided into layers. Usually, photosynthesis, respiration and transpiration are the main mechanisms of hydrogen exchange between the plants' aerial parts and the atmosphere which are taken into account in predictive models. Most often, the exchangeable OBT fraction is considered to be in equilibrium with the TFWT pool and consequently, the models do not distinguish the two fractions (Keum et al., 2006). Some models consider a possible degradation of OBT into HTO with a half-life of some days, whereas others consider there is no loss of tritium from the organic compartment when OBT is formed (Keum et al., 2006). Comparison with experimental data seems to lead to the conclusion that models considering no return routes from the OBT to the HTO pool are more suitable for dose calculation (Keum et al., 2006). In all examples found, the dose contributions due to TFWT and to OBT are treated separately in dose predictive models.

The choice of one model rather than another has to be made considering the particular situation in question. Among the different cases to be considered, it is particularly important to make the difference between a low-level but chronic exposure (for example in case of continuous release) and an acute but large release (for example due to an accidental situation).

4.1. Models for short-term exposure (dynamic equations)

4.1.1. TFWT activities

Most experimental data for tritium uptake as TFWT in plants was found to follow first-order kinetics. Exponential equations such as previously given in Eq. (2) have been fitted with these experimental data.

Another approach to describe the dynamic transfer of HTO from atmosphere to aerial parts was based on the balance of matter including tritium. The following Eq. (6) comes from the balance between inflow and outflow of the foliar compartment (Belot et al., 1984; Murphy, 1984). This equation includes the transpiration flux of tritiated water through the stomata. It has been applied in models such as RODOS (Melintescu and Galeriu, 2005).

$$\frac{dC_{\text{TFWT}}}{dt} = \frac{\gamma v_c}{M_w} (\rho_a C_a - \beta \rho_v C_{\text{TFWT}}) + \frac{v_c}{M_w} (\rho_v - \rho_a) C_{\text{sr}} \quad (6)$$

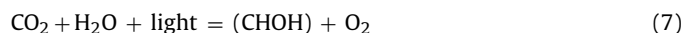
C_{TFWT} is expressed in Bq kg^{-1} ; C_a : HTO concentration in the atmosphere around the plants (Bq m^{-3}); C_{sr} : HTO concentration in the water of the rooted layer of the soil (Bq kg^{-1}); v_c : exchange velocity of water at canopy (m s^{-1}); γv_c : exchange velocity of tritiated water (m s^{-1}), $\gamma = 0.95$; ρ_v : saturated air humidity at vegetation temperature (kg m^{-3}); ρ_a : air humidity at reference level (kg m^{-3}); M_w : mobile water mass in plant leaves over a unit of soil surface (kg m^{-2}); β : inverse of the isotopic discrimination factor, $\beta = 0.91$.

Some authors also proposed models for the canopy resistance and for the soil resistance which takes place during the evaporation and the diffusion of HTO (Murphy, 1984; Melintescu and Galeriu, 2005).

4.1.2. OBT activities

OBT is produced by the photosynthesis process, from TFWT incorporated at the time of contamination, in the meteorological conditions of the moment.

The rate of water assimilation can be obtained using global stoichiometric relation of photosynthesis process (7):



The flux of newly formed non-exchangeable OBT through photosynthesis can be obtained as a product of water assimilation rate and HTO concentration in leaf water, corrected with two factors² (8). The first one is the stoichiometric factor coming from (7), this factor is 0.41 (18/44). The second is the discrimination factor for isotopic fractionation. The TRS-364 report displays some experimental values reported in the literature for this discrimination factor (*D*) in different plants (McFarlane, 1976; Garland and Ameen, 1979; Kim and Baumgärtner, 1994; IAEA, 2007b). According to experimental data, *D* ranges between 0.5 and 0.6.

$$F_{\text{OBT}} = DCP_{\text{CO}_2}C_{\text{TFWT}} \quad (8)$$

where F_{OBT} : flux of newly formed non-exchangeable OBT through photosynthesis ($\text{Bq s}^{-1} \text{m}^{-2}$); *D*: discrimination factor for isotopic fractionation; *C*: stoichiometric factor ($\text{kg}_{\text{H}_2\text{O}} \text{kg}_{\text{CO}_2}^{-1}$); P_{CO_2} : net assimilation rate of CO_2 ($\text{kg}_{\text{CO}_2} \text{m}^{-1} \text{s}^{-1}$); C_{TFWT} : concentration of HTO in plant leaves where the photosynthesis is active (Bq L^{-1}); F_{OBT} has to be integrated on the whole leaf surface and the time of water contamination.

During night, the maximum night production rate is supposed to be $670 \mu\text{g}_{\text{CO}_2} \text{m}^{-2} \text{s}^{-1}$ (per m^2 of soil, for the maximum leaf area index).

Maximum assimilation rate is about $800 \mu\text{g}_{\text{CO}_2} \text{m}^{-2} \text{s}^{-1}$ for C3 plants and $1600 \mu\text{g}_{\text{CO}_2} \text{m}^{-2} \text{s}^{-1}$ for C4 plants.

4.2. Models for chronic exposure (static equations)

The specific activity model (SA model) is commonly used to describe a situation of chronic exposure. This type of model considers the establishment of equilibrium between the different environmental compartments (atmosphere, precipitations, soil, plants, animals. . .). The SA model for tritium is formulated in terms of the tritium concentration in water. In this way, OBT concentration is calculated as the activity in the water equivalent of the dry matter (the water produced by complete combustion of the dry material) (IAEA, 2007b).

Static models proposed to predict TFWT activities in the event of chronic exposure have been derived from dynamic equations used for short-term exposure and validated using empirical data.

4.2.1. TFWT activities

- To express the TFWT concentration under steady-state conditions, the dynamic equations used for short exposures could be derivated for a very long time of exposure (Belot et al., 1979; Garland and Cox, 1982). This approach requires the introduction of some assumptions, in particular (Belot et al., 1979):

The stagnant air of the sub-stomatal cavities is considered to be saturated by water vapour;

The majority of the tissue water is accessible for rapid turnover; The diffusion of HTO is faster in the TFWT compartment than through the stomata;

Little water is translocated from the exposed leaves;

Steady-state conditions prevail.

The steady-state concentration and the uptake rate constant described in (2) could then be expressed as in (9) and (10):

$$C_{\infty} = \frac{\chi\alpha}{\rho_{\text{sat}}} \quad (9)$$

$$k = \frac{\rho_{\text{sat}}}{\alpha\mu r} \quad (10)$$

where χ : concentration of HTO vapour in air (Bq L^{-1}); ρ_{sat} : water vapour density in air saturated at the ambient temperature (g mL^{-1}); α : ratio of ^3H to ^1H in water to that in vapour at equilibrium; $\alpha = 1.1$; μ : mass of water contained per unit area of leaf (g cm^{-2}); *r*: exchange resistance for HTO (H_2O) between the leaf and the air (s cm^{-1}).

This equation was validated experimentally for grape (Belot et al., 1979) and bean (Garland and Cox, 1982) leaves. The authors confirmed that the exchange resistance for HTO and for H_2O as vapour are practically equivalent. However, it was noticed that the equation has to be modified to consider the leaf burden due to tritiated water from the soil.

- The application of (6) enables the prediction of HTO concentrations in plant leaves the equilibrium (C_{TFWT}) as a function of the concentrations in the atmosphere (C_a) and soil (C_{sr}). Such models are based on the assumptions that (IAEA, 2007b):

- the temperature of the air, the soil surface and the leaf are the same,

- the mass of plant water is constant,

- the HTO concentration at the soil surface is equal to the concentration in root water.

The simplest of such models was proposed by Belot (Belot, 1986) with the following Eq. (11):

$$C_{\text{TFWT}} = (1 - H) \times C_{\text{sr}} \times C_a \quad (11)$$

where *H* is the relative humidity of the atmosphere (L m^{-3}).

Eq. (12) uses the same parameters, based on theoretical and experimental considerations. This equation takes into account the isotopic proportions of ^3H to ^1H in water and in vapour at equilibrium through the α ratio (Belot et al., 1979; Garland and Cox, 1982; Couchat et al., 1983; Murphy, 1984; Amano and Kasai, 1988; Amano and Garten, 1991; IAEA, 2007b).

$$C_{\text{TFWT}} = \alpha(H \times \frac{C_a}{H_a} + (1 - H) \times C_{\text{sr}}) \quad (12)$$

where H_a is the absolute humidity (L m^{-3}).

In TRS 364 and in the associated technical document (TecDoc), the EMRAS group draws attention on the fact that the relative contributions of air moisture and soil water to the tritium concentration in the plant depend on the part of the plant under consideration. Eq. (12) applies specifically to plant leaves that, under normal circumstances, draw the majority of their tritium from the air. Theoretically, Eq. (12) does not apply to fruit, tubers or root crops, which draw a larger fraction of their tritium from the soil (Spencer, 1984; IAEA, 2007b). However, the equation could be used in practice for all plant parts since soil water concentrations are lower than those in air moisture in a situation of atmospheric release. Furthermore, for all the equations, the averaging time for parameter calculation (*H*, H_a , C_a , C_s) should reflect conditions that prevail very close to the considered time, since turnover of tritium in plant leaves is very fast.

² Galeriu, D., Paunescu, N., Rascob, W. Review of processes and parameter uncertainties of present tritium modeling. RODOS (WG3) – TN (98)–08. Available at <http://www.iaea.org/files/share/emras/default.asp?lg=a&fd=355>.

4.2.2. OBT activities

Since most of the dry matter of the plant is formed in the presence of water, OBT (combustion water) and TFWT concentrations should theoretically be equal. However, because of isotopic fractionation and hydrogen contents, the OBT specific activity is lower than that of HTO. The concentration of the non-exchangeable OBT is measured through the HTO activities found in water resulting from the complete oxidation of the plant organic matter. This concentration could be expressed as the tritium concentration in tissue water corrected by a discrimination factor as in Eq. (13) (Peterson and Davis, 2000; IAEA, 2007b).

$$C_{\text{OBT}} = DC_{\text{TFWT}} \quad (13)$$

where C_{OBT} : concentration of non-exchangeable OBT in the water produced by complete oxidation of the plant dry matter (Bq L^{-1}); D : discrimination factor.

The OBT concentration at harvest in plants is considered to be proportional to the mean TFWT concentration in leaves over the growing phase. This is based on the assumption that plant growth is nearly linear and that most of the non-exchangeable OBT, once formed, remains stable until harvest (IAEA, 2007b).

On the other hand, hydrogen (and thus tritium) inventories in plants vary along the growth stage. For that reason, some crop growth models created to assess the photosynthesis rate in plants have been successfully used to predict OBT formation depending on the stage of growth of the plant (Melintescu and Galeriu, 2005; Keum et al., 2006). Some of these growth models are based on a sigmoid-type function with input data including: the biomass at full development, the initial biomass and the growth rate constant (Baumgartner and Kim, 1995; Keum et al., 2006). Other models such UFOTRI and the FDMH in RODOS use more complex approaches with the influence of environmental factors.

4.2.3. Overall tritium activity in plants

The discrimination factor could be taken into account to establish the overall activity of all the tritium fractions in the plant (Guétat et al., 2008a, b) such as in (14):

$$C_{\text{total}} = [H + 0.3 \times (1 - H)] \times [(1 - H_p) \times W_{\text{eq}} \times D + H_p] \times C_{\text{a,w}} \quad (14)$$

where C_{total} : overall activity of all the tritium in the plant; H_p : proportion of water in the plant; W_{eq} : water equivalent of dry matter; D : discrimination factor between ^3H and ^1H ; $C_{\text{a,w}}$: HTO concentration in air moisture (Bq L^{-1}).

Moreover, the calculation of the OBT concentration per fresh plants weight has to consider a water equivalent factor, corresponding to the mass of water produced per mass of combusted dry matter. This leads to relation (15). The water equivalent factor is estimated according to the hydrogen content of protein, fat and carbohydrate (7%, 12% and 6.2%, respectively) and the fraction of each molecule type in the dry matter of the considered plant. For most edible plants, this factor is from 0.5 to 0.6 (IAEA, 2007b).

$$C_{\text{OBT}} = (1 - \text{WC}) \times \text{WEQ} \times D \times C_{\text{TFWT}} \quad (15)$$

where WC: fractional water content of the plant; WEQ: water equivalent factor.

4.3. Conclusions about models

Model developments and validations have been performed following experimental work and thanks to specific workshops. For instance, some models have been tested in the framework of the BIOSphere MOdel Validation Study (BIOMOVs) (Barry et al., 1999), VALidation of Model Predictions (VAMP), BIOSphere Modeling and

ASSESSment (BIOMASS) (Golubev et al., 2002b; IAEA, 2003) or Environmental Modeling for Radiation Safety (Davis et al., 2005a; IAEA, 2007b; Galeriu et al., 2008) programs. The EMRAS II program has started in 2009 and is dedicated to the testing of models for tritium from routine and accidental releases.³ Some general conclusions about predictive models for tritium have been drawn from such international intercomparisons.

First of all, models globally demonstrate a good understanding of tritium transfer into plants. However, the uncertainties linked to the models remain significant (Galeriu et al., 2008). Indeed, due to the multitude and the complexity of the phenomena involved, the models cannot be based only on theoretical assumptions but must also use empirical data as far as possible. Unfortunately, due to the difficulty to carry out experimentations in real conditions, there is a lack of experimental data to test the models. Moreover, the rare data available from experiments are specific to particular plant species and to local conditions. The generalization of the results obtained for each particular test to other cases is not trivial; for instance, experimental data on grain plants such as wheat or rice cannot be extrapolated to leaf vegetables like lettuces.

A second point is that the model performances are better for steady-state conditions than for dynamic conditions. The final report of the ^{14}C and tritium group of the EMRAS program quoted uncertainties of a factor of at least 2 for steady-state conditions, and 5 for dynamic conditions. Indeed, steady-state conditions are never reached in the real world because of the incessant fluctuations of environmental factors such as tritium activities delivered by the source, wind direction, temperature, relative humidity of air and soil. These variations are slight during more or less continuous tritium release but could be much greater with short-term release.

Comparisons of predictions given from various models for the same scenario in during workshops have highlighted the large discrepancies occurring (Barry et al., 1999; Galeriu et al., 2008). Their causes are difficult to identify because the conceptual models and the associated parameters differ in too many points. Developing a standard conceptual dynamic model for tritium dose assessment, in particular for accidental short-term releases, should be one of the objectives of future international projects (Galeriu et al., 2008).

5. Conclusions and pending questions

Releases of tritium from nuclear plants are currently low and limited to relatively few areas. Because of the potential health hazards associated to tritium, assessments of the radiological risks are required. To estimate the risks, an understanding of tritium's behaviour in the various ecological compartments is necessary. Over the last decades, the major processes of tritium transfer and cycling in the environment and the main parameters for tritium uptake and fate in plants have been described. The fate of tritium in plants has been studied with particular interest because photosynthetic plants are the basis of the ecological trophic networks, and thus, play a key role, directly or indirectly, in human nutrition. The ways of contamination, the biological mechanisms of tritium uptake and incorporation into plants and the associated kinetics have been largely detailed in the literature. Models have been established to predict the dose engaged in the event of long or short-term release. However, some unsolved questions still remain.

First, although many studies have dealt with short-term tritium exposure, only little experimental data are available on chronic exposure. This lack of data prevents the validation of the models reported in the literature, especially for plants grown in natural

³ <http://www-ns.iaea.org/projects/emras/emras2/default.htm>.

conditions. Moreover, because of the difficulty to carry out experimental studies about tritium exposure in the long term, most of the studies dealing with chronic exposure have considered only a few days or weeks (Garland and Ameen, 1979; Amano and Garten, 1991; Amano et al., 1995). The exceptions concern monitoring around nuclear facilities; in such cases, tritium concentrations in selected plants are available over several years but not all the data required for reliable models have been systematically recorded. For instance, a lot of studies about tritium in tree rings (Yamada et al., 1989; Kozak et al., 1993; Fuma and Inoue, 1995; Kalin et al., 1995; Yamada et al., 2004; Vichot et al., 2008) have been carried out; such studies give information about annual releases of nuclear facilities but do not really help understanding tritium incorporation into plants during long-term exposure. Furthermore, detailed studies about the effects of chronic tritium exposures to very low levels are needed.

More generally, additional experimental data are required, especially concerning tritium uptake and incorporation in plants at night and in rainy or snowy weather, the translocation of OBT to the different organs of plants and the rate of OBT loss after exposure (Galeriu et al., 2008).

Another gap in current knowledge concerns the biological aspects of tritium uptake and incorporation in plants. Although the qualitative and quantitative aspects at the scale of enzymatic reactions are well understood, this is not the case on the scale of whole plant metabolism. The metabolic pathways have been well described but the quantitative balances between metabolic pathways have not. The respective importance of the different phenomena is not clear. The physiological data required to quantitatively define the biological processes are currently missing. Moreover, biological measurements are particularly important to establish dose predictive models. For instance, the accuracy of such models largely depends on the overall conversion rate from TFWT into OBT, which is usually considered as constant throughout the life of plants. However, some recent studies have pointed out the differences in OBT incorporation with the growth stage (Atarashi-Andoh et al., 2002; Choi et al., 2005; Keum et al., 2006; Boyer et al., 2009). As a consequence, the quantification of these variations requires further research to improving the models.

Another point is the absence of an available OBT standard. This lack prevents the validation of an analytical method for OBT measurement and data comparison between laboratories. In fact, the interlaboratory comparison of OBT measurements has highlighted relatively large differences among the results (Workman et al., 2005). In this context, the characterization of standard reference materials, especially for environmental concentration ranges, seems to be necessary. Up to now, only Harms (Harms and Jerome, 2004) proposed the standardization of tritiated thymidine. However, such a molecule could be a reference for calibration, but is useless for general predictive dose models. Although many of the models proposed are extremely promising, owing to the lack of references, comparison between the different models remains unfruitful.

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