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Abstract

This study gives an overview on the current knowledge about the behaviour of tritium in plants and in food chains in order to evaluate the ingestion pathway modelling of existing computer codes for dose estimations. The tritium uptake and retention by plants standing at the beginning of the food chains is described. The different chemical forms of tritium, which may be released into the atmosphere (HT, HTO and tritiated organics), and incorporation of tritium into organic material of plants are considered. Uptake and metabolism of tritiated compounds in animals and man are reviewed with particular respect to organically bound tritium and its significance for dose estimations. Some basic remarks on tritium toxicity are also included. Furthermore, a choice of computer codes for dose estimations due to chronic or accidental tritium releases has been compared with respect to the ingestion pathway.

Strahlenexposition durch Tritiumfreisetzungen von NET - Datenbasis und relevante Parameter für das Verhalten von Tritium in biologischen Systemen

Zusammenfassung

Diese Studie gibt einen Überblick zum gegenwärtigen Kenntnisstand über das Verhalten von Tritium in Pflanzen und in Nahrungsketten, verbunden mit einer Bewertung der Modellierung von Ingestionswegen in verschiedenen Computercodes zur Berechnung der Dosis. Zunächst wird die Aufnahme und Retention von Tritium in Pflanzen, die am Anfang der verschiedenen Nahrungsketten stehen, beschrieben. Dabei werden die verschiedenen chemischen Formen von Tritium, die in die Atmosphäre freigesetzt werden können (HT, HTO und tritiierte organische Verbindungen), sowie die Inkorporation von Tritium in das organische Material der Pflanzen berücksichtigt. Aufnahme und Metabolismus von tritiierten Verbindungen in Tieren und im Menschen, die Bildung von organisch gebundenem Tritium und seine Bedeutung für Dosisberechnungen werden betrachtet. Eine kurze Abhandlung über Tritiumtoxizität ist ebenfalls enthalten. Schließlich wird eine Auswahl an bestehenden Computercodes zur Dosisabschätzung für Normalbetrieb oder für unfallbedingte Tritiumfreisetzungen hinsichtlich der Modellierung des Ingestionspfades verglichen.

Contents

List of Abbreviations and Notations

C_a	tritium concentration in atmospheric moisture	(Bq/ml)
C_s	tritium concentration in soil water	(Bq/ml)
C_l	tritium concentration in tissue water of leaves	(Bq/ml)
DNA	deoxyribonucleic acid	
e	vapour pressure	(mb)
E	saturated vapour pressure at temperature T	(mb)
HT	tritiated hydrogen	
HTO	tritiated water	
LAI	leaf area index	
OBT	organically bound tritium	
r_{mes}	mesophyll resistance	(s/m)
r_{st}	stomatal resistance	(s/m)
R_a	aerodynamic resistance	(s/m)
R_b	quasi-laminar boundary resistance	(s/m)
R_c	canopy or surface resistance	(s/m)
RH	relative humidity	
RNA	ribonucleic acid	
T	temperature	(°C)
TFWT	tissue free water tritium	
v_d	deposition velocity	(m/s)

Introduction

The assessment of the radiological impact of tritium releases is an essential part of the safety considerations which go in parallel with the development of fusion reactor technology. In the current stage of development it is necessary to predict the impact of probable tritium releases for licencing purposes. Mathematical models and computer codes are effective tools for this task.

Models are also necessary to estimate the contamination level after tritium releases. At definite contamination levels administrative countermeasures are considered to be required. The ICRP (ICRP 40, 1984) recommended a lower intervention level of 5 mSv that refers exclusively to the ingestion pathway , and an upper intervention level of 50 mSv for radiation exposure during the first year after an event. For comparison, the natural radiation exposure (in terms of effective dose equivalent) in the Federal Republic of Germany amounts to 2 mSv per year for man, with a variation of 1 to 6 mSv per year (BMU, 1988).

Models are based on the current understanding of the problem and on existing data that are relevant, but necessarily they simplify the real system. Sets of equations express and interpret the assumptions, that have been made about the real system, in a quantitative way.

With respect to dose consequences from tritium releases to the environment, the real system is very complex. It includes atmospheric dispersion, uptake by soil and vegetation, transport through food chains and uptake by man. The transfer rates of tritium to these different compartments depend on various parameters, which have been studied extensively in the previous time to provide a large data base.

This study only reviews the current knowledge on the tritium behaviour in biological systems after a tritium release to the atmosphere considering different chemical forms of tritium that can be released by a fusion reactor. It deals with tritium uptake by plants, standing at the beginning of several food chains, the tritium turnover in animals, the different pathways by which tritium can reach man and finally its behaviour in man, representing the end consumer.

Normally, atmospheric dispersion models calculate the radiological consequences from atmospheric tritium releases only from inhalation and skin absorption. If the ingestion pathway to man is considered additionally, tritium in plants and in particular in OBT (organically bound tritium) can become an important contribution to dose.

In practice it is not possible to measure the radiation exposure from ingestion of contaminated food. It must be calculated by means of a model. This requires the knowledge of following input parameters:

- tritium concentrations in foodstuffs originating from contaminated vegetation, which can be calculated from environmental parameters knowing the tritium concentrations in the ambient air,
- tritium concentrations in foodstuffs originating from animals that have been exposed to the tritium plume and/or were fed with tritiated feed,
- the rate of intake of different foodstuffs,
- the contaminated fraction related to the total intake of a given foodstuff and
- the dose conversion factor.

In order to estimate the dose fraction due to tritium from the ingestion pathway, dispersion models are coupled to compartment models which describe the behaviour of different chemical forms of tritium in relevant food chains.

Tritium releases to the environment can be modelled for two different situations:

1. For chronic or routine releases over extended periods of time constant atmospheric tritium concentrations and equilibrium conditions between the different compartments considered are assumed. Models based on this assumption are called static or steady state models, which base on the concept of the "specific activity model" proposed by ANSPAUGH et al. (1973).

2. After transient releases, e.g. in accidental situations, activity is rapidly lost from the local atmosphere due to dispersion, so that equilibrium assumptions are not valid. The activity concentrations of the vegetation drop rapidly after exposure to atmospheric tritium. The uptake und loss rates are highly dependent on environmental parameters and on the chemical form in which tritium was released. In this case dynamic models are used where temporal behaviour of tritium in the ecosystem under study is taken into account.

In the last chapter of this study, existing models are compared with respect to the assumptions which are made to describe the tritium uptake by plants, as the basis for ingestion models, and the ingestion pathway itself.

1 Tritium in Plants

1.1 Uptake of HTO by Plants

Concerning the uptake of HTO by plants in radioecological publications a confusion is still found, though nowadays reasonable models describing the mechanisms already exist.

From botanical point of view plant water supply in terrestrial environments occurs by uptake of soil water via the transpiration stream. During daylight, when stomata are open, considerable amounts of water are transported from the roots to the leaves and are released to the atmosphere by vapour diffusion through the stomata (transpiration). During the night, transpiration is strongly reduced because stomata of most plants are closed. Driving force for transpiration is the atmospheric demand, i.e. the difference between the vapour pressure in the atmosphere and the vapour pressure in the substomatal cavities in the leaves, which can be taken as saturized at leaf temperature. The net flux of water from the leaves into the atmosphere never can be negative. According to that conception earlier authors as CLINE (1953), expected that the HTO concentration in the leaves should approach to that in the soil water. However, such equilibration never had been observed.

From radioecological point of view HTO can enter the plant via the roots as well as via the leaves, because diffusion of the water molecules during the transpiration is not directed. Therefore this process takes place in two directions, i.e. water molecules also can penetrate from the atmosphere into the leaves. HTO ist transported along the gradient of HTO concentrations between the leaf cell surface and the atmosphere. If the HTO concentration in atmospheric moisture is higher than in soil water, the net flow of HTO may be from the atmosphere into the vegetation.

It can be expected that the equilibrium tritium concentration in the leaves is located between the tritium concentration in the soil water C_s and in the atmospheric moisture C_a. The amount of water molecules leaving the substomatal cavities corresponds to the vapour pressure in the leaves E which is saturated at given leaf temperature. The water molecules entering the leaves are proportional to the actual vapour pressure in the atmosphere e at air temperature. The ratio between influx D_2 and efflux D_1 equals the ratio between these vapour pressures. For simple estimations, the vapour pressure in stomata is assumed to be equal to the vapour saturation pressure at air temperature.

Then, this ratio is consistent to the definition of the relative humidity RH in the atmosphere:

$$D_2 / D_1 = e / E \approx RH$$

The net-transpiration T_{net} equals the difference of the two diffusion streams D_1 and D_2 which on the other hand is equal to the sapflow SF supplying the leaves with water from the soil via the xylem.

$$T_{net} = D_1 - D_2 = SF$$

This is the case if the content of tissue water in the leaves is taken as constant. An excess of transpiration however can temporary lead to water stress during the day time or chronically during a dry period.

The equilibrium tritium concentration in plant leaves C_L can be described in a simple model, which is based on the relations explained above, but neglecting extreme situations like water stress:

$$C_L = RH * C_a + (1-RH)\, C_s$$

Under equilibrium conditions the tritium concentration in the leaves results from the sum of the HTO concentrations in the atmosphere, weighted by the relative humidity, and the HTO concentration in the rooted zone of the soil weighted by the atmospheric demand (1-RH). A general assumption on which the described model is based, is a complete mixing of HTO in the leaf tissue leading to homogeneous tritium concentration and little translocation of HTO from exposed leaves.

In Figure 1.1 the main relationships of this compartment model regarding the contamination of plants by tritiated water are shown (STRACK 1989). In the figure the photosynthetic assimilation A, the respiration R and the translocation of organic molecules into other parts of the plant are included. These processes affect the turnover in the OBT compartment (organically bound tritium). The tritium transfer from the tissue free water compartment to the organic compartment (OBT) by photosynthesis is not considered in the model described above, because this transfer is much smaller than the tritium transfer from the atmospheric to the tissue water compartment. Nevertheless, the formation of OBT in plants is an important means of entry of tritium into the human food chain and therefore it should be considered in dose calculations.

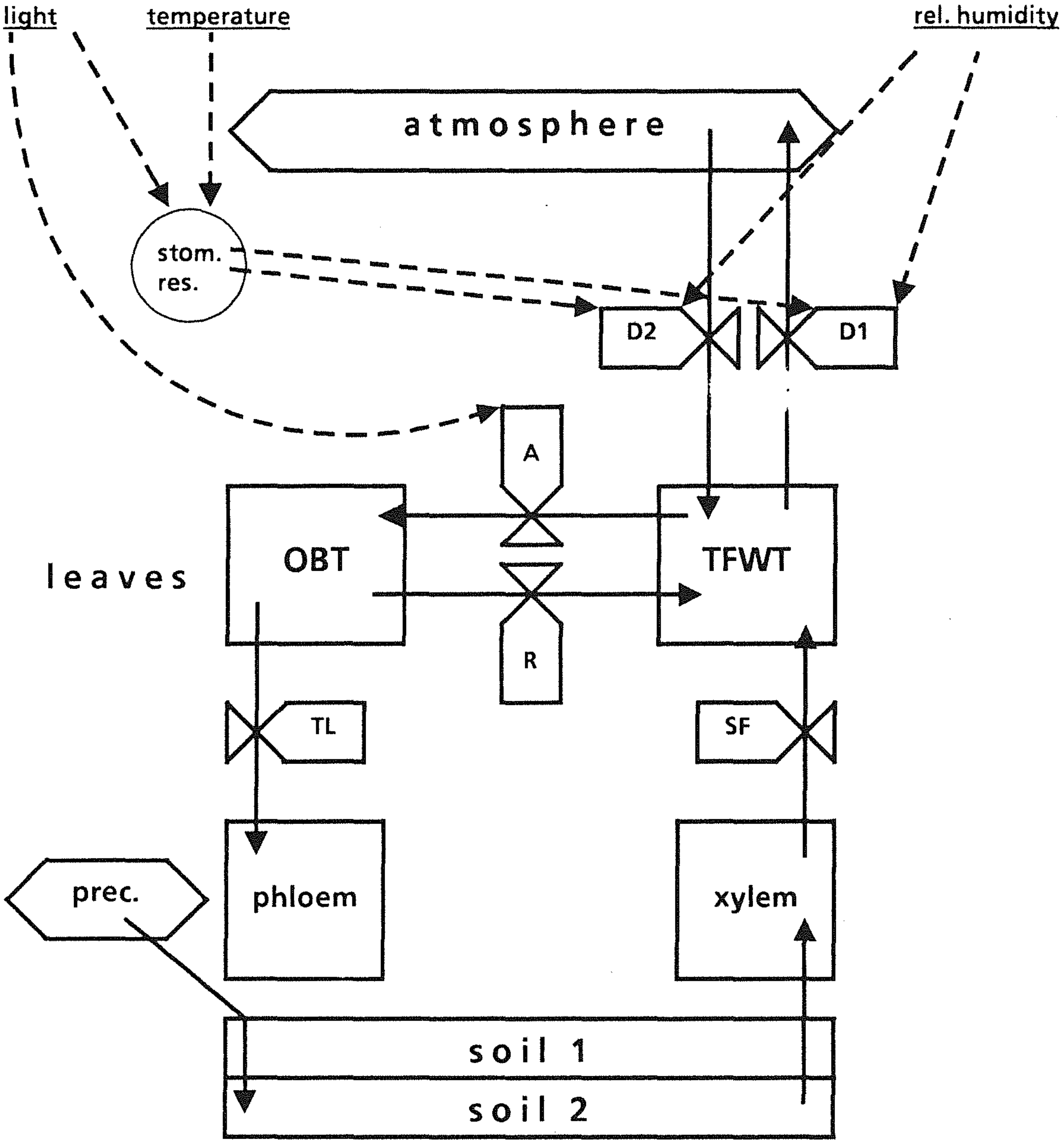

Figure 1.1: Compartment model of plant contamination by HTO with the main parameters (TFWT = tissue free water tritium compartment of the leaves, OBT = organic bound tritium compartment of the leaves, D_2/D_1 = transpirational influx and efflux of HTO controlled by the relative humidity in the atmosphere and the stomatal resistance, A = assimilation, SF = sapflow, R = respiration, TL = translocation of organic substances into different parts of the plant, prec. = precipitation, soil 1 / 2 = different soil zones)

Literature review:
To view differently the relevant literature dealing with the HTO uptake by plants it is necessary to show the development during the last decades. As already mentioned, earlier authors as CLINE described in 1953 experiments with bean plants grown in an HTO containing nutrient medium. He found out that the HTO concentration in the plant water (TFWT) does not equilibrate with the HTO concentration in the medium. Tritium concentrations approach only 45 % to 65 % that of the nutrient medium. For that he made responsible incomplete exchange with colloidally bound water in the plant tissue or a possible isotope effect. An analogous observation made RANEY and VAADIA (1965) in their experiments with sunflower and tobacco. They gave already the explanation for this observation on which recent models are based, i.e. that the equilibrium concentration depends on the actual relative humidity in the atmosphere during sampling.

KORANDA and MARTIN (1973) outlined the uptake of HTO in plants from the atmosphere, and they presented a list of experimentally determined "uptake coefficients" (HTO concentration in tissue water / HTO concentration in the atmosphere) for different kinds of plants ranging from 0.17 for black pine to 0.49 for sunflower. The authors, however, gave no information about the relative humitity in the atmosphere during sampling, that's why no further conclusions based on these values can be drawn.

BELOT et al. (1979) the first time introduced the better uptake understanding into the radioecological discussion on the environmental behaviour of tritium. In his field experiments with grape leaves exposed to tritiated vapour he observed an exponential approach of the tritium concentrations in the leaves to a predicted equilibrium value. This equilibrium concentration he defined as the product of the HTO concentration in the atmosphere C_a, the relative humidity RH, and a factor $\alpha = 1.1$, which is the quotient of the isotopic ratios T/ H in liquid and vapour (SEPALL and MASON, 1960):

$$C_L = \alpha \cdot C_a \cdot RH$$

In his model, however, BELOT not yet integrated the influence of the soil water. This was proposed by STRACK (1982) derived from observations in a systematic long-term study with a beech tree under environmental conditions. He stated that depending on which process dominates, the vapour exchange with the atmosphere or the transpiration stream from the soil, the HTO concentration of the leaf water tends to equilibrate with the atmospheric moisture or with the soil water, respectively. If the affection by both processes lies in the same range the resulting specific tritium activity of the leaves approaches the arithmetic mean of both sources. GARLAND and COX (1982) then confirmed BELOTS findings in well controlled laboratory experiments using bean leaves, and they suggested the consideration of the contribution of the soil water in the previous described way considering the relative humidity as a key parameter.

In several publications from the Savannah River Laboratory (MURPHY and PENDERGAST, 1979; MURPHY and SWEET, 1982; SWEET, MURPHY and LORENZ, 1983) studies of the dynamic environmental behaviour of tritium as HTO and HT are described. The contribution of both sources - the atmosphere as well as the soil - to the resulting tritium concentration in the leaves are taken into account in the interpretations of the observations. MURPHY the first time presented in 1984 in a detailed compendium of the relationships between HTO in air, vegetation and soil under steady state conditions a model that leads to the same consequences as it was discussed by european radioecologists. His model was more sophisticated since he takes also into account temperature differences between air and leaves (MURPHY, 1984).

This is important for the case, when the leaf temperature is higher than air temperature due to the radiant heat load, affecting the saturation vapour pressure in the stomatal cavities. The leaf/air temperature difference can be calculated from the leaf energy balance equation (RASCHKE, 1956), which can be solved for the leaf temperature for known air temperature, air-vapour pressure, radiant heat load and the diffusion resistances. Once the leaf temperature is known, the ratio of leaf saturation vapour pressure to air saturation pressure can be calculated and from that the HTO concentration of leaf water.

Steady-state modelling by the specific activity approach:
Leaf/air HTO specific activity ratios can vary greatly because complete equilibrium conditions are found rarely under realistic environmental conditions. They depend on

- relative humidity in the atmosphere
- radiant heat load
- diffusion resistances
- prevailing climate.

For dose assessments from chronic, atmospheric releases of HTO simple so called "specific activity models" were used. These models usually are based on a mean specific activity ratio expressing the overall (annual, daily etc.) average equilibration value between the tritium concentration in the atmosphere and the plants. Based on a publication from ANSPAUGH (1973) in the comprehensive NCRP REPORT No. 62 (1979) "Tritium in the environment" a ratio of 0.5 is recommended. In the US-Regulatory Guidelines (USNRG, 1977) this value is overtaken. MURPHY (1984) recommended on basis of environmental observations in the vicinity of the Savannah River Plant (humid climate) a ratio of 0.8. In the German Regulatory Guidelines (BMI, 1979) a ratio of 1.0 is included.

Dynamic modelling by the resistance model:

The diffusion of HTO from the atmosphere into tissue water of plant leaves can also be modelled by a deposition velocity v_d, which can be determined by experiments (inferential methode) or calculated from given meteorological and plant data. The deposition velocity (or conductance) is the reciprocal of leaf diffusive resistance, consisting of three components (HICKS et al., 1987):

$$v_d = 1 / (R_a + R_b + R_c)$$

The resistance component is introduced to account for the chemical and biological processes that control HTO uptake. The three components of the total leaf resistance are:

1. The aerodynamic resistance R_a, determined by atmospheric properties.
2. The quasi-laminar boundary layer resistance R_b, to account for molecular diffusivity in the vicinity of the surface.
3. The canopy resistance R_c, that combines the consequences of all uptake processes, which are affected by biological factors, into a single number.

R_a and R_b can be calculated from meteorological data, whereas for calculation of R_c also plant specific data are necessary.

Terminology:

a. Resistances associated with biological pathways (via stomata for HTO vapour) are quantified in terms of a biological resistance to transfer expressed as the effective resistances per unit area of foliar surface.
b. In the context of the "big-leaf" model, the integrated consequences of these resistances expressed per unit horizontal area of the earth's surface are quantified.

One link between these two is the leaf area index (LAI), and its equivalents that refer to other components of the biomass. The symbol r is used to identify resistances expressed in terms of a unit area of foliar surface, i.e. biological resistances.

The canopy resistance R_c is then calculated by the formula:

$$R_c = (r_{st} + s \cdot r_{mes}) / LAI$$

r_{st} is the stomatal resistance and r_{mes} the mesophyll resistance, which has to be multiplied by the solubility factor s of the gas. In the case of HTO vapour, the mesophyll resistance is neglectable.

Transfer through stomata takes place by diffusion. Stomatal resistance depends on the incident photosynthetically active radiation and varies with plant species. The effects of humidity, water stress and temperature on stomatal resistances can be taken into account by correction factors, which can be calculated from meteorological data for known plant species.

Increasing atmospheric vapour pressure deficit that corresponds with decreasing relative humidity reduces the stomatal conductance proportionally.

Stomatal resistance is dependent of plant water stress, when the leaf potential drops below a threshold value. Below this threshold value, stomatal resistance increases rapidly.

The species dependent higher and lower temperature extremes at which stomata no longer open (typically 40 °C and 5 °C, respectively), can also be taken into account for calculations of stomatal resistances. The temperature at which stomatal exchange is optimized is typically 25 °C.

At this time, R_c can be estimated from measurements of photosynthetic active radiation (PAR), air temperature, and humidity, coupled with observations of vegetative species, leaf area index and surface wetness. Effects of water stress remain to be addressed (HICKS et al., 1987).

Since the stomatal resistance is a strong function of radiation, and therefore affected by shading by other foliage, a layer-by-layer computation of net overall resistances is appropriate for a canopy with large LAI, e.g. for forests.

Uptake and retention kinetics of HTO in plants:
The uptake of tritium from atmospheric moisture into tissue water of leaves is a rapid process reaching its equilibrium level after some hours under daylight conditions when stomata are open. For instance, KLINE and STEWARD (1974) found a half-time for uptake of tritium in grass leaves of 1.1 hours and in grass stems of 32.8 hours.

The loss of tritium after end of exposure is also rapid, but several investigators observed a second and a third compartment in the excretion curves of tritium (Table 1.1). The excretion rates depend on plant species, length of exposure and the same parameters affecting stomatal resistance (light, humidity, temperature and others). During dark conditions, the retention half-time of tritium in grass leaves was found to be 6 times higher than under light conditions (KLINE and STEWARD, 1974).

The fast component represents over 90 % of the total incorporated tritium. The second, much slower component is probably attributed to organically bound tritium. For the third component the authors believed that it is from tritium in soil water.

plant	exposure time (h)	biological half-time (h)			reference
		1. component	2. component	3. component	
Fiddleneck	0.5	0.7	24		1
Burclover	0.5	0.8	25		1
Burclover	1.0	0.9	17	270	2
Wine grape	6.0 (autumn)	0.6-2.0			3
Wine grape	6.0 (spring)	0.3-0.6			3
Wine grape	4.0	0.5	30	80	4

1: ANSPAUGH et al., 1973
2: KORANDA and MARTIN, 1973
3: BELOT et al., 1979
4: GUENOT and BELOT, 1984

Table 1.1: Biological half-times of tritium in vegetation after exposure to HTO vapour under daylight conditions.

By contrast to this, HTO concentrations in leaves decline slower following an HT exposure of the plant-soil-system because of deeper diffusion of HT into the soil. After conversion to HTO in the soil, tritium is longer available to plants (SWEET et al., 1983, KIRCHMANN et al., 1986).

1.2 Uptake of HT and Tritiated Organic Compounds by Plants

When atmospheric HT comes into contact with soil it is rapidly converted to HTO by an enzyme mediated reaction. The responsible enzyme called hydrogenase is capable of reversibly activating molecular hydrogen and is present in many bacteria of various taxonomic groups, in many eucaryotic algae, in some protozoan microorganisms, and in several mosses (ADAMS et al., 1981). The enzyme also occurs outside of living organisms, but it is only active in the presence of sufficient amounts of soil water (FÖRSTEL, 1986). The process of HT deposition on soil and conversion to HTO is well studied and considered to be the main route of contamination of the environment following a release of HT (DJERASSI and LESIGNE, 1988; BURNHAM et al., 1988).

There are some reports on hydrogen metabolism in higher plants demonstrating the occurrence of a hydrogenase system. Because there is no evolutionary barrier to the expression of hydrogenase, it was suggested, that a latent, normally inactive hydrogenase may be present in many plants. RENWICK et al. (1964) found that several seedlings evolved small amounts of H_2 under sterile conditions when subjected to anaerobic stress indicating the existence of hydrogenase in these organisms. But it was not possible to demonstrate the reverse reaction, H_2 uptake. TORRES et al. (1986) found similar results with seedlings of maize, wheat and barley. They demonstrated, that the hydrogenase activity is induced by the plant itself and not by contaminant microorganisms. The hydrogenase was selectively induced in roots and to a lower extent in hypocotyls, but not in leaves (TORRES et al., 1986). Those organs can be exposed to low levels of oxygen during plant development, which can cause anaerobic stress. Other biochemical mechanisms than hydrogenase action metabolizing molecular hydrogen in plants are not known.

The deposition of HT on vegetation is generally neglected in model calculations, because it is much slower compared to soil. Values of HT deposition velocity to soil are in the range of 10^{-4} to 10^{-3} m / s, whereas deposition velocities of HT to vegetation are about two orders of magnitude lower.

Plants	Leaf area index	Deposition velocity of HT (m / s)	Reference
Exposure chamber experiments			
scots pine	?	$4\cdot10^{-7}$	Garland and Cox, 1980
loblolly pine needles	?	$1.2\cdot10^{-7}$	Sweet and Murphy, 1984
white pine	12.0	$1.2\cdot10^{-6}$	Spencer and Dunstall, 1986
trembling aspen	5.3	$1.7\cdot10^{-6}$	"
speckled alder	5.5	$1.6\cdot10^{-6}$	"
soybean	5.6	$3.7\cdot10^{-6}$	"
corn	7.6	$2.9\cdot10^{-6}$	"
Field experiments			
soybean (potted)	5.6	$4\cdot10^{-7}$	Spencer et al., 1988
corn (potted)	7.6	$6\cdot10^{-8}$	"
white spruce (potted)	12	$2.3\cdot10^{-8}$	"
red maple (potted)	?	$7.3\cdot10^{-8}$	"
red pine (naturally grown)	?	$1.2\text{-}6.0\cdot10^{-7}$	"
red maple (naturally grown)	?	$1.3\text{-}5.1\cdot10^{-7}$	"

Table 1.2: Deposition velocities of HT to vegetation calculated on a leaf area basis for tissue free water tritium (TFWT).

The values for HT deposition to vegetation are in general expressed on a leaf area basis for a better comparison to data from soil. Vegetation enhances the area for deposition of HT by several times compared to soil, depending on plant species, age and morphology. Data from field and exposure chamber experiments are listed in Table 1.2.

Short term releases of HT to the environment (field experiments in France in 1986 and in Canada in 1987) showed, that plant contamination is mainly caused by the uptake of tritium in the form of HTO which was formed from HT by oxidation in the soil. The tritium concentrations in the free tissue water depend on the relative concentrations of HTO in the soil and atmosphere. In the first hours after the release, HTO was rapidly reemitted from soil to atmosphere followed by exchange of HTO vapour with foliar water (DIABATÉ and HONIG, 1988; SPENCER et al., 1988). Later on the uptake of HTO from soil by plant roots dominated (SPENCER et al., 1988). This indicates that the upper soil layers were the major source of tritium in plants. The incorporation of tritium into organic matter was very slow and was mainly attributed to HTO. A direct incorporation of HT was not detectable during these field experiments.

McFARLANE (1978) suggested a more direct transfer of tritium when plants were exposed to HT. He compared the specific activity ratios between HTO in air and in lettuce leaves and found a higher ratio of 0.84 after HT exposure than after HTO vapour exposure (0.34). This may have been caused by a more available source (e.g. higher HTO concentration near the leaves than in the bulk air) or it may indicate that HT penetrated the plant and was converted in the substomatal cavity to HTO and subsequently absorbed from that side (McFARLANE, 1978).

Moreover, SWEET and MURPHY (1984) demonstrated that HT is also involved in some processes leading to the synthesis of organic matter. They exposed living pine needles to HT under controlled conditions in light and in dark. The deposition velocity of HT to tissue free water tritium was $1.2 \cdot 10^{-7}$ m / s in light and $0.2 \cdot 10^{-7}$ m / s in dark and was not dependent on the HT concentration. The deposition velocity to nonexchangeable organically bound tritium was two orders of magnitude lower, but higher in dark than in light. This process depends on the HT concentration suggesting that it is limited by the number of active sites such as an enzyme mediated reaction. No incorporation into organic matter was observed in soil and litter.

The specific activity of HT is higher than that in HTO vapour in the atmosphere but the absolute amount of molecular hydrogen is much smaller compared to that of water vapour. If there is a direct incorporation of HT into organic matter, it is reasonable to expect, that the specific tritium activity of organic matter synthesized from HT is greater than that synthesized from tritiated water.

Another explanation is that HT is oxidized to tritiated water and establishes a high local concentration of HTO at the site of photosynthesis and the synthesis of organic matter is faster than HTO equilibration with the rest of the cellular water. Decreased water flux in the dark and the consequent slower HTO dilution may then explain the higher production of tritiated compounds in the dark (SWEET and MURPHY, 1984).

DUNSTALL et al. (1985) made similar observations. During HT exposure over 30 days the average OBT specific activities of poplar and tomato foliage were several times greater than respective values for tissue-free water tritium (TFWT). This suggests the possibility of direct HT incorporation into OBT as proposed by SWEET and MURPHY (1984). However, DUNSTALL et al. did not exclude the possibility of contamination of OBT samples during analysis as another reason for higher OBT values.

Although vegetation is not an important sink for HT deposition to the terrestrial environment, the process of direct incorporation of HT to OBT may be important for a forest area where the needle surface can be 10-20 times that of the soil. In addition, higher wind speeds in the free canopy than at the soil surface will increase the HT deposition velocity (SWEET and MURPHY, 1981). Because of the slow decomposition rate of needle litter, OBT remains in the soil for many years.

The contribution of direct HT to OBT conversion by vegetation to the dose under chronic release situations was estimated by MURPHY (1986). Depending on the amount of atmospheric hydrogen that penetrates directly into organic matter of vegetation, he calculates doses for HT releases up to 40 % compared to tritium exposure as HTO. Also, if no direct conversion of HT to OBT is taken into account, he still calculates a dose of 4 % compared to a "pure" HTO exposure because of tritium transport through the food chain in the form of organic substances.

Another possible way of HT entry into plants, which is discussed, is the oxidation of HT to HTO by microorganisms living in symbiosis with root cells (root nodules of leguminoses). These bacteria are able to fix molecular nitrogen of the air. Some of them posess an enzyme, the so called "uptake hydrogenase", which is able to oxidize molecular hydrogen to water (EISBRENNER and EVANS, 1983). This possible way of HT uptake is not yet studied sufficiently. SPENCER and DUNSTALL (1986) found no significant difference in deposition velocities of HT to soybean (leguminose) and other plant species.

Comparison of HT and HTO uptake by plants:
KIRCHMANN et al. (1986) describe an experiment in which naturally growing potatoe, maize and rye plants were exposed to HT or HTO vapour for four hours in a closed plastic greenhouse. The main differences in soil and plant contamination following these two exposure situations were as follows:

- Initial OBT values in leaves were about 200 Bq / g per GBq released for HTO vapour and about 50 Bq / g per GBq released for HT (exchangeable OBT was not removed).
- The easier penetration of HT into the soil and the longer retention than HTO cause a longer tritium availibility after HT exposure than after HTO vapour exposure.
- The half-life of OBT in leaves was about 10 days after exposure to HTO. The loss of OBT after HT exposure was slower, which can be attributed to the longer tritium availibility in the soil.

Behaviour of tritiated organic compounds in plants:
Tritium releases during operation of the tritium processing area at Savannah River Laboratories (SRL) consist to about 1 % of tritiated organic compounds. Most of the organic tritium released was postulated to be in the form of tritiated methane (MILHAM and BONI, 1976).

There exist only a few papers on the transfer of tritium in the organic form to vegetation. MASON et al. (1973) observed that alfalfa plants incorporate tritium from tritiated methane into the plant free-water and into the organic phase. Their data indicate that in transpiring plants the incorporation of tritiated methane into the leaf free water reaches steady state in about 18 minutes. The deposition velocity of tritiated methane was only 4 % of that of HTO vapour.

1.3 Incorporation of Tritium into Organic Material of Plants

HTO is rapidly distributed throughout living organisms which contain on average 70-90 % of weight as free water. Finally, tritium can enter any hydrogen position in organic matter. The incorporation of tritium into biological molecules (e.g. proteins, carbohydrates, fats or nucleic acids) can proceed via the following processes:

a. Exchange of tritium from HTO with labile protons of organic matter. These are hydrogen atoms attached to oxygen, nitrogen or sulfur atoms. The fraction of organically bound tritium in these positions is called "exchangeable" tritium.
b. Incorporation of tritium into stable bonds to carbon atoms via metabolic processes. This fraction is called "nonexchangeable" tritium and in most references it is associated with the term OBT (organically bound tritium).
c. MATHUR-DE VRÉ and BINET (1984) note, that a third process, namely the hydration of biological macromolecules has to be considered. The hydration water of biological macromolecules represents an integral part of the macromolecular structures and plays a crucial role in maintaining the structural and conformational integrity of all biostructures. The binding to organic molecules is relatively strong due to electrostatic interactions.

Exchangeable tritium: Tritium atoms from tritiated water and exchangeable hydrogen atoms at accessible positions of organic molecules equilibrate rapidly within a few seconds. By contrary, hydrogen atoms at internal sites of coiled macromolecules exchange very slowly due to their inaccessibility to cellular water. In edible parts of green vegetables about 30 % of the organically bound hydrogen are theoretically able to exchange with tritium in free water (DIABATÉ, unpublished results). This value varies with the fat content of the biological material, because fat has a lower level of exchangeable hydrogen. The parts of exchangeable hydrogen of the main components of biological material are listed in Table 1.2.

	Total hydrogen content (in weight-% of dry matter)	Portion of exchangeable hydrogen (in weight-% of total hydrogen)
proteins	about 6.8	about 25
carbohydrates*	5.3 - 6.8	20 - 42
lipids	10 - 13	0 - 5

*in dependence of the degree of polymerization

Table 1.2: Exchangeable hydrogen content of the main components of organic material estimated from stoichiometric calculations (DIABATÉ, unpublished results).

Nonexchangeable tritium: Tritium is incorporated in carbon-bound sites of organic matter via metabolic reactions. Quantitatively, the most important process of de novo biosynthesis of biological material from water and carbon dioxide is the photosynthesis of higher plants. Furthermore, tritiated water might be incorporated into stable carbon bonds by some reactions of the glycolytic scheme and the tricarboxylic acid cycle producing tritiated intermediary products. Some of these products lead to the biosynthesis of lipides, proteins, polysaccharides and nucleic acids. Some basic enzymatic steps of the early biosynthetic incorporation of tritium were outlined by SMITH and TAYLOR (1969).

The rate of incorporation of tritium is dependent on the turnover rate of the molecule species to which it is bound. For instance, tritium is only incorporated into the genetic material of those cells that synthesize DNA, that means deviding cells. In embryonic organisms nearly all cells are dividing. With further development the dividing cells are located at special sites. Since the rate of removal of tritium is also dependent on the turnover rate of the labeled molecule, tritium bound to DNA constitutes a long-term hazard in cells with long life span.

Proteins and other biopolymeres are being continously synthesized in a biosystem. Therefore, the incorporation rate is higher.

Isotopic effects:
A widely known isotopic effect is the discrimination of tritium during the cleavage of the O-H-bond of tritiated water. This effect causes that less tritium atoms enter organic molecules during metabolic reactions compared to hydrogen atoms. By contrary tritium once fixed to carbon is cleaved more slowely than C-H-bonds.

The relative incorporation of tritium compared to hydrogen is expressed as the ratio (in some papers R value) between the specific tritium activity per gramm hydrogen of one compound or of total organic material and the specific tritium activity per gramm hydrogen in HTO administered to an organism.

Long-term exposures of Chlorella algae to tritiated water showed that the tritium incorporation compared with hydrogen incorporation into metabolites is highly variable (KANAZAWA et al., 1972). Highest ratios (around 1) were found in compounds related to the tricarboxylic acid cycle (intermediates and related amino acids), and low ratios (0.5-0.7) in primary products of photosynthesis, lipids and some amino acids. The results suggest that, once incorporated, tritium is preferentially retained during oxidative reactions (KANAZAWA et al., 1972).

An explanation to this phenomenon is given by RAMBECK and BASSHAM (1973). The increased ratios for the tricarboxylic acid cycle intermediates and related amino acids can be accounted for specific isotopic effects of several enzyme mediated steps. These effects may increase the tritium activity either in the substrate or in the product of an enzymatic reaction, depending on wether the discrimination is within the molecule (intramolecular case) or between molecules (intermolecular case). If a hydrogen atom is removed from a methyl group (intramolecular case), the activity of the reaction product will increase, because the C-H-bond is preferentially broken. When the hydrogen atom is removed from a unique position (intermolecular case), the activity of the substrate before the discriminating step will increase, because the non-labeled molecule is preferred.

In the tricarboxylic acid cycle occur three dehydrogenase reactions where an increased tritium activity of the substance before the reaction step can be expected. The highest ratio of 1.8 was found for citrate in blue-green algae. But citrate and related products represent only a small part compared to those compounds, where tritium is discriminated during synthesis. Therefore it is unlikely that enrichment of tritium could occur, even in the food chain where organic material is transported from one organism to another (RAMBECK and BASSHAM, 1973).

STRACK et al. (1979) showed for the planctonic algae culture under dynamic conditions how the ratio becomes greater than 1 in a continuous culture. They also reported a value of $I = 0.8$, where I is the ratio of $(\mathrm{OBT})_{\mathrm{actual}}$ to $(\mathrm{OBT})_{\mathrm{theoretical}}$.

OBT/TFWT ratio in environmental samples:
In some model calculations an OBT/TFWT ratio of 1 is utilized representing a conservative assumption neglecting the isotopic discrimination. This is only valid for equilibrium situations, where OBT has essentially the same specific activity as the water from which it is formed.

But there exist several evidences for an OBT/TFWT ratio greater than 1 for environmental samples and foodstuffs. Examples from all over the world are outlined in a review by BROWN (1988 a).

The reasons for this phenomenon have to be differentiated. The tritium content in the free tissue water and exchangeable OBT of an environmental sample reflects the tritium concentration in environmental water at the time of sampling due to the rapide equilibration time of tritium. The rate of tritium incorporation into nonexchangeable OBT is much more slowly, depending first of all on the exposure time and furthermore on many other parameters like temperature, light conditions, and plant species. In the natural environment it is nearly impossible to reach an equilibrium with nonexchangeable OBT, since continuous exposures to an enhanced constant tritium concentration do not occur in practice. After the end of exposure to tritium, the decomposition of OBT and formation of HTO is much more slowly than HTO loss in the plant. Therefore, an OBT/TFWT ratio greater than 1 in environmental samples can particularly be attributed to the much longer retention time of OBT compared to HTO. The specific tritium activity in OBT can exceed that of TFWT at a distinct time after the end of tritium exposure (for example BROWN et al., 1988 b).

Another explanation for enhanced OBT/TFWT ratios in environmental samples is in discussion: a direct incorporation of HT or tritiated organic compounds, e.g. methane, into OBT (SWEET and MURPHY, 1984; BELOT, 1986). The mechanism of this process is still unknown.

KIM and BAUMGÄRTNER (1986) brought the possibility of tritium enrichment in OBT by sample preparation, in particular by lyophilization, into discussion. Because of the low temperature in the sample during water removal by lyophilization, the isotope effect is high and can lead to a tritium enrichment in the residual moisture of the sample which cannot be removed. This water equilibrates with exchangeable hydrogen atoms in organic substances and can result in an apparent higher OBT value (7 - 20 %, depending on the combustion methode, thermal or plasma oxidation). However, such an effect does not completely explain the observed OBT/TFWT ratios, which should be discussed further on.

SPENCER (1984) the first time found an OBT/TFWT ratio greater than unity (1,92) in green fruits of tomato plants after continuous exposure to atmospheric HTO under controlled environmental conditions. Fruits have little exchange to atmospheric moisture, hence they maintain a low TFWT value. However, the fruit incorporates photosynthetic products translocated from the foliage. In this case OBT can exceed TFWT. This observation is not explained by a tritium enrichment mechanism, but by different contamination of leaves and fruits by atmospheric HTO.

Relevance of time of exposure:
The knowledge of tritium concentration in edible plant organs is necessary for dose estimations. The distribution of tritium in different plant organs is dependent on the time of exposure and the time of harvest. Differences are due to different metabolic activities in organs and tissues during the life cycle of the plant.

ARAI et al. (1985) studied the incorporation of tritium into organic material of edible parts of soybean, wheat and rice as a function of time of administration by a single irrigation and time of harvest. Seeds of soybean had the highest radioactivity when they were exposed to HTO at the beginning of August, the time of ripening of soybean, followed by harvest 2-3 weeks later. The incorporation of tritium into the organic material of wheat fruit was highest when HTO was administered two weeks after flowering, the period where growth of the fruit is very rapid. In rice, the highest OBT content of the fruit was found, when HTO was given two weeks after flowering and harvesting 50 days later, at the natural time of harvest. OBT concentration in the fruit portion of the rice grain was about 10 times of that found in stems and leaves.

INDEKA (1981) made similar observations with edible parts of tomato, potato, sunflower, maize and wheat as experimental plants, which were exposed to atmospheric HTO during different stages of plant development. Tritium exposure in the time after flowering resulted in higher incorporation into organic material of edible parts of plants. This shows clearly the relevance of the time of exposure for estimations of the dose due to incorporation of OBT from vegetable foodstuffs.

2 Tritium in Animals

2.1 Uptake and Metabolism of Tritium

HTO uptake:
Terrestrial animals can incorporate tritium via ingestion of tritiated water in drinking water or in feed, via inhalation of atmospheric moisture or diffusion of HTO vapour through the skin. The tissue free water tritium (TFWT) in animals quickly responds to HTO exposure. To a small extent tritium is incorporated into organic material by metabolic processes, depending on the time of exposure. If tritium reaches the animal only by one pathway, TFWT never exceeds the tritium concentration in the contaminated water. It is diluted by water of lower tritium concentration from other intake routes.

Tritium, the radioactive isotope of hydrogen, can be incorporated into all organic molecules by substitution of ordinary hydrogen and follows essentially the same biological pathway. Its entry into an organism and its metabolism is a complex interaction of a number of parameters, like for ordinary water or feed.

Because of the short range of the tritium β-radiation only the intracellular components need to be considered for possible health effects. A typical distribution of hydrogen in animal cells is shown in Table 2.1 for rat liver cells as an example (COMMERFORD, 1984). More than three quarters of the total hydrogen are in the form of water and about 14 % are in proteins. Less than 0,5 % of the hydrogen are in nucleic acids (DNA = deoxyribonucleic acid, RNA = ribonucleic acid).

cellular component	weight - % of the cellular component	% of total hydrogen
water	69.0	77.4
proteins	21.0	14.0
polysaccarides	3.8	2.4
lipids and steroids	5.0	5.8
RNA	1.0	0.3
DNA	0.2	0.07

Table 2.1: Distribution of hydrogen in rat liver cells (from COMMERFORD, 1984).

Tritium can enter into all these components, but by different mechanisms. HTO enters the water pool within minutes after its entry and is distributed uniformly by the blood stream. This is also valid for the exchangeable part of organic hydrogen (hydrogen attached to nitrogen, oxygen or sulfur atoms) (see also chapter 1.3). For this reason tritium in tissue water and in exchangeable positions of organic matter are considered together as a single component. Tritium incorporation into non-exchangeable hydrogen positions (hydrogen bound to carbon atoms) is only possible by enzymatically catalyzed reactions.

The typical distribution of exchangeable and non-exchangeable hydrogen in animal cells is shown in Table 2.2 for rat liver cell as an example (COMMERFORD, 1984). It can be seen that 95 % of the exchangeable hydrogen are present in water and only 5 % are in organic matter. Less than 20 % of the total cellular hydrogen is non-exchangeable, mainly in proteins, and only about 0,3 % is non-exchangeable in nucleic acids.

cellular component	weight -% of exchangeable H related total cellular H	weight -% of non-exchangeable H related to total cellular H
water	76.6	0
proteins	3.0	10.9
polysaccarids	0.7	1.6
lipids and steroids	0.07	5.8
RNA	0.09	0.2
DNA	0.01	0.06

Table 2.2: Distribution of exchangeable and non-exchangeable hydrogen in rat liver cells (from COMMERFORD, 1984).

If the hydrogen distribution of the whole body is compared to the hydrogen distribution in the cell, about two third of the total hydrogen occur in the exchangeable and about one third in the non-exchangeable hydrogen compartment (CLAUSEN, 1980).The incorporation of tritium into these compartments depends on whether tritium is taken up as HTO or OBT, on the duration of tritium exposure, and on the anabolic and catabolic activity of the organism.

OBT uptake:
An important pathway for the intake of tritium by animals is the ingestion of feed in which tritium occurs organically bound. A fraction of ingested and absorbed OBT will be oxidized to HTO for production of energy. This part will enter the TFWT-pool of the animal. The other fraction of absorbed OBT will be incorporated into animal's tissue. OBT of animal tissue responds faster in the case of OBT ingestion in comparison to HTO ingestion and reaches higher levels than TFWT. In this case, an OBT/TFWT ratio greater than 1 occurs. But this is no accumulation effect or enrichment against a concentration gradient, e. g. like iodine accumulation in the thyroid gland. There is no depot of hydrogen in any organism.

In most models describing the tritium turnover in animals it is assumed, that all OBT from food is oxidized metabolically to HTO after it is absorbed in the gastrointestinal tract. However, this does not correctly reflect the real situation.

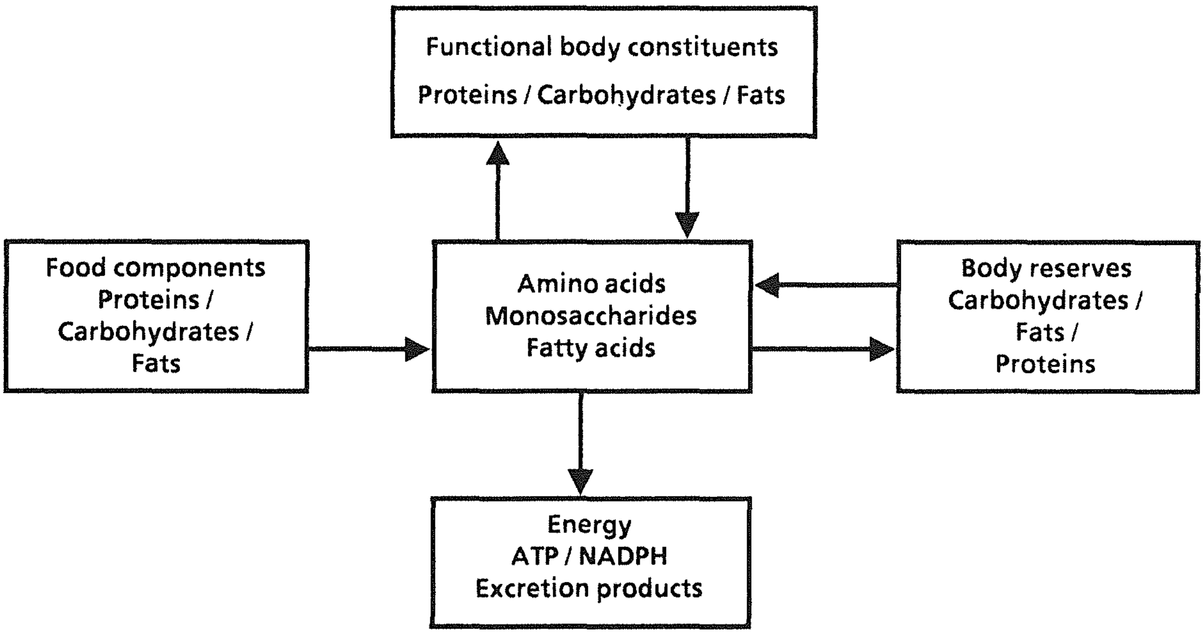

Figure 2.1: The fate of organic food components (from VAN DEN HOEK, 1986).

Figure 2.1 shows the pathways of ingested foodstuffs (VAN DEN HOEK, 1986). The food, consisting of high molecular proteins, carbohydrates and fats as main components and other molecules like nucleic acids (only about 1 % by weight of dry matter), is digested in the gastrointestinal tract. This is essentially a process of hydrolytic cleavage, resulting in smaller molecules like amino acids, monosaccharides, fatty acids and nucleotids. These molecules can enter the blood stream, by which they are transported to metabolyzing organs for following processes:

1. Production of energy: This is metabolic oxidation of OBT producing molecules rich in energy (ATP), and HTO, which will enter the body water pool.
2. Synthesis of functional body constituents: e. g. enzymes, hormones, structural elements and secretion products, e. g. milk. Many of these compounds are proteins. This process results in a conversion of ingested OBT into an other type of OBT.
3. Synthesis of body reserves, particularly fat: This represents also conversion of one type of OBT into another.

2.2 The Lactating Cow as Experimental Animal

An important pathway of tritium to man is via the classic grass-cow-milk pathway. Therefore, the lactating cow is often used as experimental animal. Milk synthesis takes place in the mammary gland and involves continuous formation of protein (casein), fat (milk fat) and carbohydrate (lactose). Incorporation of tritium into these components can be studied easily and for a long time and can be related to the chemical form in which tritium is ingested by the animal (VAN DEN HOEK et al., 1983 and 1985).

However, one has to bear in mind, that it is not allowable to compare the results from cow experiments (ruminants) with those from monogastric animals, because the digestion is different, particularly for carbohydrates.

Differences in tritium metabolism are also between lactating and non-lactating cows (e. g. beef cattle). The water turnover is higher in lactating cows, producing about 20 litres milk per day.

Possibilities of modelling:
A convenient way of describing the transport of tritium from the diet of a lactating cow into her milk is the usage of a transfer coefficient or transfer rate (LASSEY, 1980). Under steady-state conditions it is defined as the fractional rate of transfer from diet to milk and is expressed as a percentage of the daily dietary intake, secreted per litre of milk.

The validity of this concept to the transfer of tritium from drinking water depends on the dilution of the body water pool with non-tritiated water from external and/or internal sources. This may be water from ingested food, from metabolic processes in the organism and from exchange processes through the respiratory tract (inhalation) and through the skin (skin absorption).

The rate of incorporation can also be estimated by comparing the specific activities between the component and its precursor. In the case of milk components, the precursor is tissue water, which can be measured by urine analysis. It is generally accepted that the tritium content of tissue water is similar to that in urine water. This procedure takes into account all dilution processes with non-tritiated water and therefore it is the best estimate of tritium incorporation for steady-state conditions.

Since more than 95 % of tritium in milk is in the water phase (average water content of milk is 87 %), it is also possible to express the transfer of tritium as a ratio of the specific activities in milk water and drinking water.

2.2.1 Transfer of HTO from Drinking Water to Milk

When cows received a constant daily dose of tritiated water, steady-state concentrations in milk water, milk fat, lactose and casein were reached in about 18 days (VAN DEN HOEK et al., 1983). Table 2.3 lists the transfer rates that have been observed for cows in full lactation phase, on a diet with very low water content (hay and concentrate) and with a daily water consumption of nearly 50 l.

milk constituent	transfer rate
milk water	1.52
lactose	0.026
milk fat	0.026
casein	0.009

Table 2.3: Transfer rates for HTO to milk constituents (in % of the daily tritium intake, secreted in one litre of milk) (VAN DEN HOEK et al., 1983).

Specific activity ratios:

In the same experiment, the specific activity ratios between the tritium concentrations in milk constituents and in drinking water were determined for equilibrium conditions (see Table 2.4).

milk constituent	specific activity ratio
milk water	0.83
lactose	0.48
milk fat	0.30
casein	0.22

Table 2.4: Specific activity ratios in milk constituents during continuous HTO administration to lactating cows related to drinking water (VAN DEN HOEK et al., 1983).

The value of 0.83 for milk water can be considered as the upper limit that can occur under practical circumstances, because of the low water contents of the feed material used during the experiments.

When the specific activity ratios of milk constituents are related to tissue water, the following values are obtained as shown in Table 2.5.

milk constituent	specific activity ratio
milk water	1.00
lactose	0.58
milk fat	0.36
casein	0.28

Table 2.5: Specific activity ratios of milk constituents during continuous HTO administration related to tissue water (VAN DEN HOEK et al., 1983).

2.2.2 Transfer of OBT from Feed to Milk

The transfer of OBT from feed to milk has been determined by administration of tritiated hay, containing homogeneously labeled OBT, to lactating cows (VAN DEN HOEK, 1985). The results show (see Table 2.6) that part of the OBT has been converted to HTO during catabolism and entered the body water pool. About one half of the tritium secreted daily into milk can be attributed to the milk water fraction and the other to the transfer to organic material in milk.

milk constituent	transfer rates
total milk	1.60
milk water	0.84
milk fat	0.53
casein	0.18
lactose	0.04

Table 2.6: Transfer rates from OBT to milk for equilibrium conditions (in % of daily ingested OBT secreted in 1 litre of milk at steady-state) (VAN DEN HOEK et al., 1985).

The ratios of specific tritium activities between milk constituents and hay (Table 2.7) clearly demonstrate the importance of OBT for the incorporation of tritium into milk fat and casein.

milk constituent	specific activity ratio
milk water	0.10
milk fat	0.84
casein	0.49
lactose	0.05

Table 2.7: Specific activity ratios between milk constituents and ingested OBT during continuous OBT administration (VAN DEN HOEK et al., 1985).

The high values of the specific activity ratio in milk fat suggest, that most precursors are of similar specific activity as the ingested hay. The low values for lactose indicate, that most tritium atoms in food are replaced by hydrogen during the fermentation process of the food in the forestomachs of the ruminant animal and during following metabolic pathways leading to lactose.

The average tritium levels in milk constituents reached a plateau after about 20 days, indicating that a semi-steady state condition was obtained. During this period, tritium levels were highest in milk fat. After the end of OBT exposure, tritium levels in casein became higher and remained so during the lactation period.

2.3 Distribution of OBT in animal organs

The incorporation of tritium into organic matter of various organs of animals was studied after either HTO or OBT administration by different workers (PIETRZAK-FLIS et al., 1978, ROCHALSKA and SZOT, 1979, KIRCHMANN et al., 1977). Since organs or tissues differ in their metabolic activity, it is consistent to compare the specific activities in the non-exchangeable fraction of OBT (tritium bound to carbon). Tritium can enter or release these positions only by enzyme-mediated reactions. An example is given in Table 2.8.

tissue	specific activity of non-exchangeable OBT (kBq/g H)		ratio of specific activities (OBT/HTO)
	OBT	HTO	
brain	6.97	4.18	1.67
liver	23.89	3.61	6.62
kidney	22.66	3.34	6.78
lung	21.45	3.66	5.86
spleen	19.06	3.17	6.01
heart	14.21	2.18	6.52
ovary	18.28	3.55	5.15
testes	20.97	5.76	3.64
small intestine	17.70	2.79	6.34
muscle	11.27	2.40	4.70
skin	5.87	1.82	3.23
femur (cortial)	1.44	0.71	2.03
bone marrow	9.09	1.05	8.66
adrenal gland	12.96	2.59	5.00
adipose tissue	6.37	1.44	4.42

Table 2.8: Specific activity of non-exchangeable OBT in rabbit tissues after HTO or OBT administration (dried alphalpha) for 4 weeks. The daily dose was 185 Bq of HTO or OBT per gram of body weight (from PIETRZAK-FLIS et al., 1978).

After OBT administration, the highest concentrations were found in liver and kidney, representing organs with high metabolic activity. The specific activities after HTO administration were highest in brain and testes (only one sample). Lowest concentrations were found for both modes of tritium administration in organs with low metabolic activity like adipose tissue, muscle and bone.

KIRCHMANN et al. (1977) studied the tritium distribution in various organs of calves and pigs comparing the total incorporation into organic matter of main organs relative to the ingested tritium activity (Table 2.9 with pig as an example). The values of total incorporation rates vary depending on the mass of the organs considered. After HTO administration, about 0.7 % of total activity ingested was incorporated into organic matter of the main organs of the pig. 4.08 % and 10.98 % of total activity were incorporated, when tritium was ingested as tritiated milk powder or tritiated potatoes, respectively. That means that the incorporation from tritiated OBT was 5.8 - 15.7 times higher than from HTO depending on food. In calves, the incorporation from tritiated milk powder was 15.1 times higher than from HTO.

Organ	incorporation rates of H-3 (in % of total activity ingested)		
	tritiated water	tritiated milk powder	tritiated potatoes
lungs	0.019	0.111	0.2
liver	0.04	0.098	0.447
spleen	0.0045	0.016	0.036
pancreas	0.0031	0.023	0.032
duodenum	0.0086	0.086	0.186
large intestine	0.019	0.042*	0.278
kidneys	0.0056	0.047	0.08
muscles	0.6	3.17	9.66
brain	0.0023	0.01	0.023
thymus	0.0019	0.027	0.036
sum	0.7	4.08	10.98

*contamination by faeces?

Table 2.9: Distribution of tritium in organic fractions of pig organs after administration of HTO, tritiated milk powder or tritiated potatoes for 4 weeks. The analyzed organs represent 49 % of weight of the whole animal (from KIRCHMANN et al., 1977).

PIETRZAK-FLIS et al. (1978) found that the incorporation of tritium into organic components of rabbits exposed to tritium in food is higher by factor 1.66 - 8.66 than in rabbits exposed to HTO (Table 2.8). For rats, ROCHALSKA and SZOT (1979) found 2.5 - 15.5 times higher incorporation after exposure to tritiated food.

2.4 Contribution of OBT to the Dose

Tritium incorporation from HTO into organic matter of tissues and higher incorporation rates after OBT ingestion compared to HTO uptake have consequences on the radiation dose, since OBT has a longer retention time in the body than HTO. The turnover of tritium in the body is usually expressed as the biological half-life, which varies with the size of the body water compartment of the species regarded, the age and the climatic conditions. On cessation of exposure, HTO of the body decreases much more rapidly than OBT. OBT in body tissues will be converted into smaller mobile molecules, which may be oxidized to HTO or re-used for synthesis of new organic molecules. Both tritiated compounds, water and organics, may be secreted or stored.

The decrease of the specific activity of tritium in tissue water after cessation of tritium exposure can be described by an equation with three exponential terms. For instance for mice which had been exposed to a constant level of HTO since conception, the following equation was found for the specific activity A in tissue water at time t in Bq/ml (COMMERFORD, 1984):

$$A(t) = 4.47 \times 10^{6} e^{-0.44t} + 5890 e^{-0.03t} + 380 e^{-0.0041t}$$

The three terms indicate that tritium was present in at least three compartments which had biological half-lives of 1,6, 23 and 170 days. The relative size of these compartments can be calculated by dividing the coefficient of each term by the value of its exponent. The result is that 97 % of the tritium in these compartments represented tritium in the form of water (half-life 1,6 days), 2 and 1 % were organically bound with a half-life of 23 and 170 days, respectively.

TAKADA and KASIDA (1979) found, that the retention of OBT was high in those tissues in which the initial tritium concentration was relatively low, such as fat tissue, brain and muscle. This suggests that the excretion of OBT is largely determined by metabolic turnover of the tissue constituents, which is considered to be slow in these organs.

When half-times of tritiated water in various animal species are compared, a correlation between body weight and rate of water turnover can be observed (YOUSEF, 1973). The biological half-lives of tritium in different species are compared in Table 2.10.

species	biological half-lives (days)	
	first compartment	second compartment
mouse	1.1 - 1,6	23
kangaroo rat	13.3	114
chicken	4.6	
goat (lactating)	2.9 - 5.3	
goat (non-lactating)	6.7 - 10.4	
pig	3.8 - 4.3	
cow (lactating)	3.1 - 4.0	33
cow (non-lactating)	4.0	40

Table 2.10: Comparison of biological half-lifes of tritium in body water after administration of HTO (reviewed by VAN DEN HOEK et al., 1983).

Half-times in milk components:

In cows, a fraction of ingested tritium (HTO or OBT) is incorporated into fat and protein molecules, which are deposited in certain tissues of the body. This fraction can be reutilized at a later time either for catabolism or for resynthesis of milk components. An enhanced remobilization of body reserves occurs after the birth of a calf.

The half-times of tritium in milk components after chronic HTO exposure, determined by VAN DEN HOEK et al. (1983), show also two compartments (Table 2.11).

milk component	biological half-lives (days)	
	first compartment	second compartment
milk water	3.6	43.7
lactose	3.6	43.7
milk fat	4.1	224.8
casein	4.3	24.3

Table 2.11: Half-lives of tritium in mik components after chronic administration of HTO to a lactating cow (VAN DEN HOEK et al., 1983).

The slow compartments represent only 4 % of the total tritium in milk under conditions of continuous HTO uptake and less than 0.1 % after a single dose.

The rapid decrease of tritium in milk water reflects the high rate of water turnover in the lactating cow. The long lived component for milk fat probably represents tritium release from body fat.

The contribution from OBT to the dose after prolonged exposure to various tritium sources compared to the dose from tritium in body water were reviewed by MYERS and JOHNSON (1987) (see also Table 2.12).

The dose increases by about 4-8 %, when OBT formation in the body is considered additionally after tritium ingestion as HTO. Only ETNIER et al. (1984) found a 70 % increase by model calculations. The enhancement of dose can reach up to 100 %, if all organic materials in the normal diet are tritiated. The estimated increase of 400 % for the case, that all C-bound hydrogen atoms in proteins are replaced by tritium, may not be relevant in practice because it is not likely that the only source of tritium would be tritiated protein in the diet.

tritium source in diet	% increase in dose above that due to tritium in body water	reference
HTO in drinking water	4 - 8	PIETRZAK-FLIS et al., 1982; TAKEDA and KASIDA, 1979; KIRCHMANN et al., 1977
tritium in dried vegetable material (dried alfalfa given to rabbits)	40 - 100	PIETRZAK-FLIS et al., 1978
tritium in fresh vegetables containing 80 % HTO and 30 % OBT in equilibrium (calculated from values for dried vegetable material and for HTO)	16 - 35	
tritium in seed and desert items	40	MARTIN and KORANDA, 1972
tritium in dried milk powder, fed to calves and pigs	50 - 120	KIRCHMANN et al., 1977
tritium in all organic material in a normal diet	100	COMMERFORD, 1984
all C-bound hydrogen atoms in proteins are replaced by tritium	350 - 400	COMMERFORD, 1983; ETNIER et al. 1984

Table 2.12: Approximate contributions to the dose from OBT formation in the body from different tritium sources in the diet after prolonged exposure, additionally to the dose due to tritium in body water alone (from MYERS and JOHNSON, 1984).

2.5 Is there Tritium Enrichment in the Food Chain?

Since there is no evidence for a concentration process for tritium either in plants nor in animals, no tritium enrichment should occur in the food chain. Long term experiments have shown, that tritium from HTO fails to enter all nonexchangeable positions of hydrogen (VAN DEN HOEK, 1979). In addition, tritium concentrations in various organic molecules are not equal even under equilibrium conditions (COMMERFORD et al., 1977; ROCHALSKA, 1977).

The ratios of the specific tritium activities in organic material of animal tissues or secretion products and either in tissue water or tritiated compound ingested also indicate, that there is no tritium enrichment in different animal species after continuous tritium intake (Table 2.13)

animal	tritiated compound ingested	specific activity ratio	reference
rats	HTO	0.2 - 0.3*	THOMPSON and BALLOU, 1956
rats	HTO over three generations	0.25 - 0.47*	LASKEY, 1973
rats	tritiated meat over three generations	0.22 - 0.37**	PIETRZAK-FLIS et al., 1982
mice (liver and testes)	HTO	0.25 - 0.4*	HATCH and MAZRIMAS, 1972
mice (globine, glycogen, DNA)	HTO	0.25; 0.55; 0.75	COMMERFORD et al., 1977
kangaroo rats	living in a tritiated environment	1.2 - 1.6*	MARTIN and KORANDA, 1972
calves	HTO	0.19 - 0.25**	KIRCHMANN et al., 1977
calves	tritiated milk powder	0.16 - 0.44**	KIRCHMANN et al., 1977
pigs	HTO	0.10 - 0.15**	KIRCHMANN et al., 1977
pigs	tritiated milk powder	0.08 - 0.31**	KIRCHMANN et al., 1977
rabbits	constant tritium specific activity ratios in feed and drinking water through three generations	0.95 - 1.0*	MOGHISSI et. al., 1987
cow (lactose, casein, fat of milk)	tritiated hay	0,05; 0,49; 0,84**	VAN DEN HOEK et al., 1985
goat (protein and fat of milk)	tritiated hay	0,29-0,3; 0,37-0,52**	VAN DEN HOEK, 1986

*ratio of specific activities between OBT in tissue and TFWT.
**ratio of specific activities between OBT in tissue and the tritiated compound ingested

Table 2.13: Specific activity ratios in animals after chronic tritium administration.

Only in kangaroo rats the specific activity ratios of OBT in tissue water and TFWT were greater than 1 due to isotopic effects of tritium involved in enzymatic reactions or in transfer across liquid-vapour phases (MARTIN and KORANDA, 1972). Kangaroo rats lived in a generally tritiated environment at Sedan Crater created by a nuclear explosion in 1962 in a desert of southern Nevada. Many generations of those animals have been chronically exposed to tritium from conception to maturity by drinking water and food. Moreover, they have a unique water metabolism which allows them to maintain a positive water balance only eating dry grain.

In laboratory experiments under defined exposure conditions, OBT in animal tissue never exceeded the concentration of administered HTO and/or OBT at steady-state equilibrium conditions. These results demonstrate, that there is no tritium enrichment in the food chain.

3 Tritium in Man

3.1 Tritium Toxicity

The biological toxicity of tritium is mainly due to the β-particle emitted in the course of transmutation to He-3. The β-particle produces ions and excites molecules along its tract with a path length of an average of less then 0.9 µm in biological material. In addition, the tritium decay to helium-3 leaves an organic molecule that is positively charged. This may be reactive in causing molecular alterations. Quantitatively, however, biological effects of this transmutation are small in comparison to those caused by the track of the β-ray (FEINENDEGEN et al., 1980 a).

Because of the very short range of the tritium β-particle in biological matter, radiation effects are limited to the subcellular vicinity of the tritium location and vary with the radiosensitivity of the biological structure. Sensitive and critical targets are in particular cell nuclei, harbouring the radiosensitive macromolecule DNA. By contrast, the cytoplasm of the cell is less sensitive to radiation from tritium.

Stem and germ cells are considered to be the most radiosensitive cell types. Stem cells, which are distributed over the whole body, have the capability of self renewal and produce differentiated functional cells. Radiation to the nuclei of these cells can induce late somatic effects, e. g. cancer. Radiation damage to germ cells in ovary or testes can furthermore induce genetic effects, e. g. mutations in the offspring. The rate of tritium incorporation into such sensitive sites, the turnover of the sensitive site itsself and the tritium position at the molecular and submolecular level need to be taken into consideration for calculation of the dose equivalent of incorporated tritium (FEINENDEGEN, 1967).

Biological manifestation of tritium toxicity may be compared directly with effects of other types of radiation in terms of "relative biological effectiveness" (RBE). The RBE value is largely a question of reference radiation. It can also vary with dose and dose rate, as well as with the biological endpoint, e.g. cancer induction or genetic effects.

For determining the RBE values for tritium, experimental animals are exposed to HTO and the biological effects are compared to those, which were induced by an appropriate reference radiation. FEINENDEGEN (1980 b) found, that the tritium β-spectrum is comparable to the spectrum of Compton- and photoelectrons generated in matter by 100 kV X-rays. For this type of reference radiation, the RBE of tritium was determined to be 1. Other workers found variations in RBE values for tritium between 1 and 3 using Co-60 gamma rays or 250 kV X-rays as reference (NCRP 63, 1979, ICRU 40, 1986). These studies yielded no evidence that biological effects induced by incorporated tritium differ in any qualitative characteristic from those induced by external X- or gamma radiation.

It is, however, obvious that the RBE value determined for HTO is not valid for other tritiated compounds. An extreme example is the comparison of toxicity of tritiated water and H-3-thymidine, a nucleotide precursor. It has been estimated that tritiated thymidine injected intravenously into mice results in an exposure of stem cell nuclei that is 50 times greater than from a single administration of the same activity given as tritiated water, because of its selective incorporation into DNA. For oral uptake of H-3-thymidine, the factor ist only 10 (FEINENDEGEN et al., 1980 b).

The term RBE is restricted to radiobiology and it should be distinguished from the quality factor (Q) that is employed in radiation protection, where Q is related to RBE and LET (linear energy transfer). The International Commission on Radiological Protection (ICRP 26, 1977) has adopted the quality factor of 1 for tritium β-rays for radiation protection purposes.

In 1986, the Joint Task Group of the ICRP and the ICRU (International Commission on Radiation Units and Measurements) stated, that the dependence of biological effectiveness of a radiation dose upon the intracellular distribution of energy should be considered in dose calculations. The distribution of energy, which is determined from the type of the charged particles and their velocity, is specified by two physical quantities, the linear energy transfer (LET) and the lineal energy, y.

LET is the quotient of the energy lost by a charged particle in matter and the distance, which is traversed by the particle. y is the quotient between the energy imparted to the matter in a volume of interest by an energy deposition event and the mean chord length in that volume. y was introduced, because it is more related to biological effectiveness than LET. For radiation protection purposes it was recommended, that radiation quality should be based on lineal energy in a 1 µm diameter sphere of ICRU tissue (muscle).

Based on this relation, the effective quality factor for tritium β-particles is 2 (ICRU 40, 1986).
It should be noted that the quality factor does not include the effect of tritium, that has been ingested as OBT. To account for OBT, the dose due to tritiated water has to be multiplied by a modifying factor which can be different for several types of food (see also chapter 2.4).

3.2 Entry of Tritium

Tritium can enter the body by inhalation, absorption through skin and by ingestion. The degree of incorporation and retention of tritium in the body is effected by the chemical form of the tritiated compound which is taken up.

Exposure to HT mainly results in a dose to the lung by inhalation due to the low solubility of hydrogen gas in water. Only a small fraction (0.004 %) of inhaled HT dissolves in the blood and is transported throughout the body (PETERMAN, 1982, PETERMAN et al., 1985). Part of it is converted to HTO, presumably by hydrogenase-containing bacteria in the gastrointestinal tract and behaves like tritium incorporated as HTO. After exposure, dissolved HT is exhaled.

HTO vapour is taken up by inhalation as well as by absorption through skin. The uptake by inhalation is about twice as much when persons are active during the exposure. When persons are at rest, the intake is approximately equal along the two routes. In the body, HTO diffuses freely and rapidly across all cell membranes and equilibrates with body fluids within minutes. When the skin is splashed with tritiated water, intake through the wetted skin occurs. Tritiated water soaks into the extravascular layer of the skin and subsequently diffuses into the body (PINSON and LANGHAM, 1957; OSBORNE, 1968).

Tritium can be ingested from drinking water and/or from food containing HTO and OBT. The nonexchangeable part of OBT will enhance the dose in comparison to tritiated water, depending on the composition of the diet. Food rich in proteins will have another effect than food rich in carbohydrates.

3.3 Behaviour of Tritium in the Body

The residence time of tritium in the body is one of the major factors affecting the health aspects because the toxicity is directly proportional to the residence time (ICRP 30, 1979). The disappearance of tritium activity from the organic and water compartment of the human body was investigated by measuring of tritium in urine after accidental exposures to tritium. The results were interpreted by a model dividing the total hydrogen of the human body into three compartments with different half times (REINIG and SANDERS, 1968; MOGHISSI et al., 1971 and 1972; BENNET, 1973). These half times are considered to be in the ranges of 8-10, 20-36 and 200-550 days (BENNETT, 1973).

According to CLAUSEN (1980), 67 % of the human hydrogen occur in the free tissue water and in exchangeable positions of the organic matter with a half-life of approximately 10 days. 3 % of hydrogen disappear with a half-life of 30 days (hydrogen in immobile water and in labile carbon bonds) and about 30 % disappear with a half-life of 1 year (stabile hydrogen, bound to carbon). In animal experiments, where HTO or OBT was fed to calves or pigs for 4 weeks (KIRCHMANN et al., 1977, see chapter 2.3), it was shown that less than 1 % of ingested HTO was incorporated into organic fractions representing the hydrogen compartment with the long half-life, which amounts to about one third of the total hydrogen in the body. When tritium was administered as tritiated food, 4 - 11 % of the ingested tritium activity was incorporated into this compartment.

With regard to genetic consequences of human exposure to tritium it is of general interest to evaluate the incorporation of tritium into DNA of the most radiation-sensitive organs like ovary or testes. For a very rough estimation of tritium incorporation into these critical organs, a uniform tritium concentration in non-exchangeable OBT can be assumed, although it is known, that tritium concentrations vary in different organs (Table 2.8, chapter 2.3). Assuming that the critical organ has a wet weight of several gram and that it represents about 0,01 % of the total body weight of an adult, only 0,001 % of tritium ingested as tritiated food will be incorporated into non-exchangeable OBT of the critical organ. If it is furthermore assumed that the DNA content is 0,2 % of the organ's wet weight (value for liver DNA, COMMERFORD, 1984), 0,000002 % of tritium ingested as tritiated food will be incorporated into the DNA of the critical organ.

However, this estimation should be considered critically since it is known that new DNA is primarily synthesized by proliferating cells which hence incorporate tritium into non-exchangeable hydrogen positions of DNA. Rapidly proliferating cells are rare in adults (bone marrow, testes) but common in juveniles and embryos. Once incorporated into DNA, tritium remains there until the cell's death. This is the reason for the long half-time of tritium in DNA. For instance, tritium in the DNA of mice liver cells decreases with a half-time of 318 days (COMMERFORD et al., 1982). When cells with longer life span than liver cells, e.g. primary oocytes, are exposed to tritium during proliferation, the incorporation of tritium into non-exchangeable positions of DNA becomes of importance, since tritium may remain in these positions over several decades. However, the probability of tritium incorporation into these positions is very low, because oocytes only proliferate during the fetal development (SLATKIN and COMMERFORD, 1982).

For the purpose of dose estimations, the turnover and metabolic behaviour of tritium is described by several models which are briefly described.

ICRP model:

In the ICRP recommendation "Limits for Intakes of Radionuclides by Workers" (ICRP 30, 1979) the Annual Limit of Intake (ALI) of tritium is calculated on the basis of the water balance model for Reference Man (ICRP 23, 1975) only considering the retention of tritium in body water. The model assumes that the daily intake of water by fluids, water in food, and water obtained by metabolic oxidation of food (total 3000 ml per day) is equal to daily water losses. The biological half-life of tritium in the free water pool is assumed to be 10 days. In practice, this value can vary due to personal habits and ambient temperature within a range of 4 - 18 days. The formation of OBT from tritiated water is neglected since it is thought that the uncertainty in dose due to individual variations in retention of body water is much greater than the error introduced by ignoring OBT. Intake of tritiated organic compounds is also not taken into account.

The new ICRP recommendation "Age dependent Doses to Members of the Public from Intake of Radionuclides: Part 1" (ICRP 56, 1990) additionally considers the formation of OBT after ingestion of tritiated water and suggests higher dose conversion factors for the ingestion of OBT.

For dose calculations after intake of tritiated water, a uniform distribution of HTO and OBT in the body is assumed. For the retention of tritium, which can be represented by a three-component exponential function (SNYDER et al., 1968, SANDERS and REINIG, 1968), a simplified two-component exponential function is applied in the ICRP recommendation:

$$R(t) = 0{,}97\ e^{-0{,}693t/T1} + 0{,}03\ e^{-0{,}693t/T2}$$

The half-life of the first component (T1) is assumed to be 10 days in adults, which can be calculated from the daily water intake and the mass of the body water. The daily carbon balance and the mass of the total body carbon are used to calculate the half-life of the second component (T2), representing the OBT-compartment. For adults, a half-life of 40 days is taken. The half-times for both components are lower for children in dependence on their age. The dose contribution from OBT is assumed to be 10 %.

In the dose calculations for intake of OBT it is assumed that all OBT containing food components are digestible and can be absorbed by the gastrointestinal tract. Indigestable food components (e.g. fibres) are assumed to represent only a small proportion of the total OBT and are neglected.

The incorporation rate into non-exchangeable OBT after HTO intake was assumed to be 1 - 5 % of the tritiated water entering the blood (TAKEDA and KASIDA, 1979:1 - 5 % of the total tritium in the wet tissues of rats were found in the non-exchangeable OBT). After ingestion of OBT, the non-exchangeable OBT-fraction was about nine times higher than after HTO ingestion (PIETRZAK-FLIS et al., 1978, ROCHALSKA et al., 1977). Because of the conservative assumption that 50 % of the total ingested OBT-activity is incorporated into non-exchangeable OBT of tissues, the following model for tritium retention from OBT is proposed by the ICRP:

$$R(t) = 0{,}5\ e^{-0{,}693t/T1} + 0{,}5\ e^{-0{,}693t/T2}$$

This means, that 50 % are assumed to show the same behaviour as tritiated water (T1 = 10 days for adults) and the remaining 50 % are considered to follow the metabolic behaviour of carbon (T2 = 40 days for adults). The half-times for children are lower depending on age.

The derived dose coefficients (or dose conversion factors) derived from the biokinetic data for ingestion or for inhalation of HTO and OBT in dependence of age are also listed in ICRP Publication 56 (1990). For OBT, the dose coefficients are higher by a factor 1,8 to 2,8 (Table 3.1).

age at intake	effective dose equivalent in Sv/Bq for		factor for OBT
	HTO	OBT	
3 months	$5{,}5 \cdot 10^{-11}$	$1{,}0 \cdot 10^{-10}$	1,8
1 year	$4{,}1 \cdot 10^{-11}$	$1{,}1 \cdot 10^{-10}$	2,7
5 years	$2{,}6 \cdot 10^{-11}$	$6{,}7 \cdot 10^{-11}$	2,6
10 years	$1{,}9 \cdot 10^{-11}$	$5{,}4 \cdot 10^{-11}$	2,8
15 years	$1{,}6 \cdot 10^{-11}$	$4{,}0 \cdot 10^{-11}$	2,5
adult	$1{,}6 \cdot 10^{-11}$	$4{,}0 \cdot 10^{-11}$	2,5

Table 3.1: H-3-dose coefficients (effective dose equivalent in Sv/Bq to age 70 years) for ingestion or inhalation of tritiated water and OBT (according ICRP 56, 1990).

NCRP model:

The National Council on Radiation Protection and Measurements (NCRP 62, 1979) utilizes a three-compartment model proposed by BENNETT (1973). A water balance of 3.0 l/day and retention half-time components of 9.6, 30 and 450 days for tritium in the body (see Figure 3.1) are assumed.

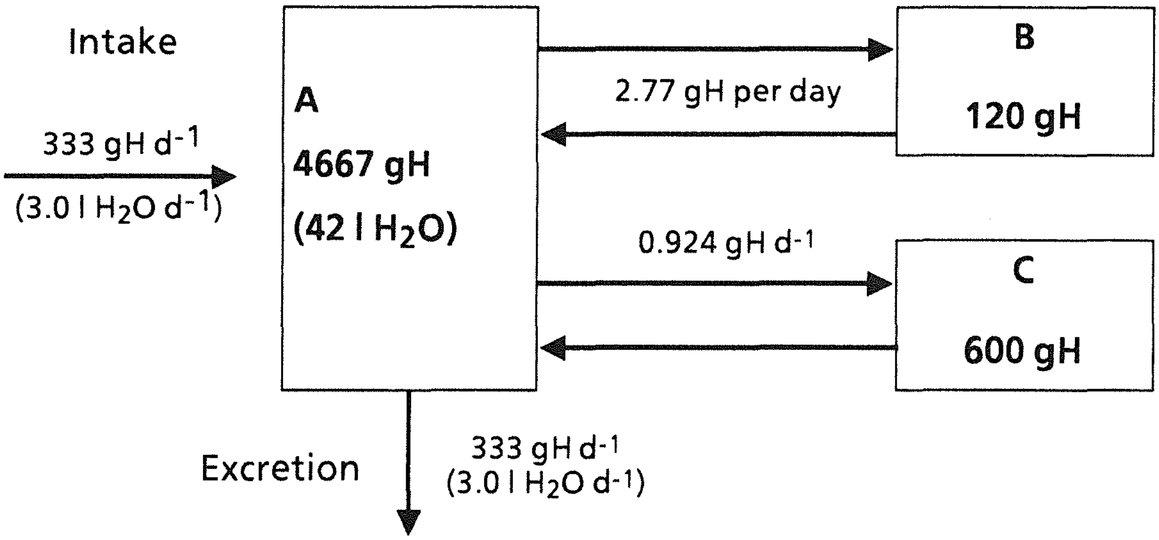

Figure 3.1: Three-compartment model of hydrogen in the body (according NCRP 62, 1979).

Human data are based on those for the ICRP Reference Man (ICRP 23, 1975). Compartment A represents the hydrogen pool in the body water (60 % of the body mass) with an average half time of 9.7 days. Accounting for transfer to the bound compartments reduces this value to 9.6 days. The hydrogen of dry tissue matter is distributed between compartment B (30 days half-time) and compartment C (450 days half-time). The different half-times reflect the differences in chemical bonding of hydrogen in tissue. The model assumes that all ingested tritium, HTO and OBT, enters directly into the free water compartment. The NCRP indicates, that the formation of OBT from HTO may be accounted for by multiplying the free body water dose by a factor of 1.2.

This model is useful for dose calculation due to intake of tritiated water by ingestion, inhalation and absorption through skin considering the formation of OBT. The uptake of tritiated organic compounds by food is not taken into account.

ETNIER, TRAVIS and HETRIC model:
The four-compartment model proposed by ETNIER et al. (1984) accounts for conversion of dietary OBT to OBT in the body. The total hydrogen of the human body is devided into the free body water compartment A including the exchangeable hydrogen of organic molecules, two organic compartments which include adipose tissue mass (B and D), and a rapidly metabolizing ("fast fat") compartment C, which represents a small highly active subcomponent of adipose tissue (see Figure 3.2).

The model includes following assumptions:

- The daily hydrogen intake is 346 g according the water balance of 3.0 l / day (ICRP 23, 1975), and arises to 91 % from free water and to 9 % from organically bound hydrogen in food.
- Intakes of tritium as free water result in 100 % uptake into compartment A. A part is used for metabolic reactions supplying organically bound tritium.
- A portion of organically bound hydrogen in food (input into C) is oxidized immediately during the metabolism supplying energy and enters the free body water compartment A. The authors assume that 43 % of the daily carbohydrate intake and 20 % of the daily fat intake go along this way. The remainder is stored in adipose tissue (compartment D) until needed.
- Approximately 25 % of ingested proteins are stored as fat (input into C) and 75 % are utilized for the building up of cellular components (input into B).

- The removal half-times of the organic compartments B and D (22 days and 450 days) represent degradation and turnover of general structural and functional proteins and other macromolecules within the body. This turnover is known to vary with age and between organs and tissues. Longest half-times have been reported for DNA.
- The half-time for excretion of body water is assumed to be 10 days.

ETNIER et al. conclude that the total radiation dose from tritiated water exposure alone increases by the factor 1.7 when the incorporation of tritium into organic substances of the body is taken into consideration. When organically bound tritium of food is ingested and assimilated in the bound compartments of tissues, the total body dose can increase by a factor of up to 4.5, depending on the bound-to-loose ratio of tritium in the diet.

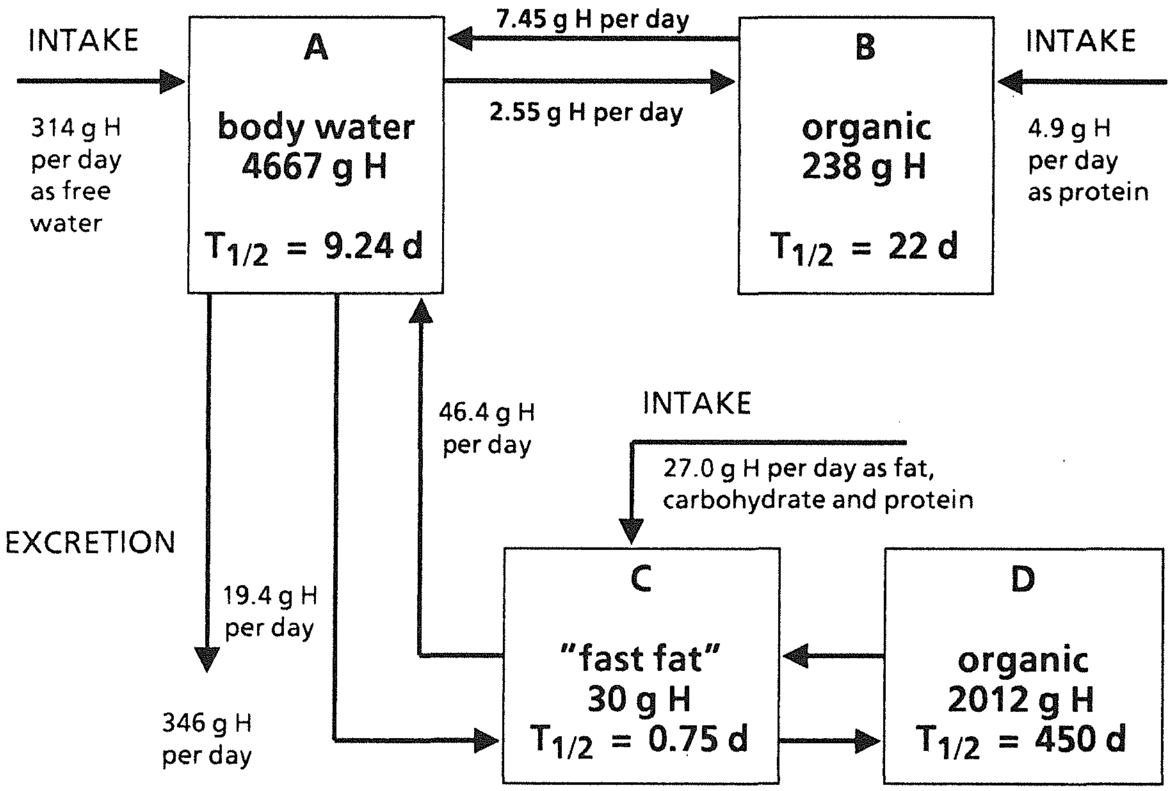

Figure 3.2: Four-compartment model of hydrogen metabolism according to ETNIER et al. (1984).

KILLOUGH model:

KILLOUGH (1982) proposes a five-compartment model utilizing the physiological assumption for the ICRP's Reference Man (ICRP 23, 1975). He adds two additional compartments for cortical and trabecular bone (see Figure 3.3).

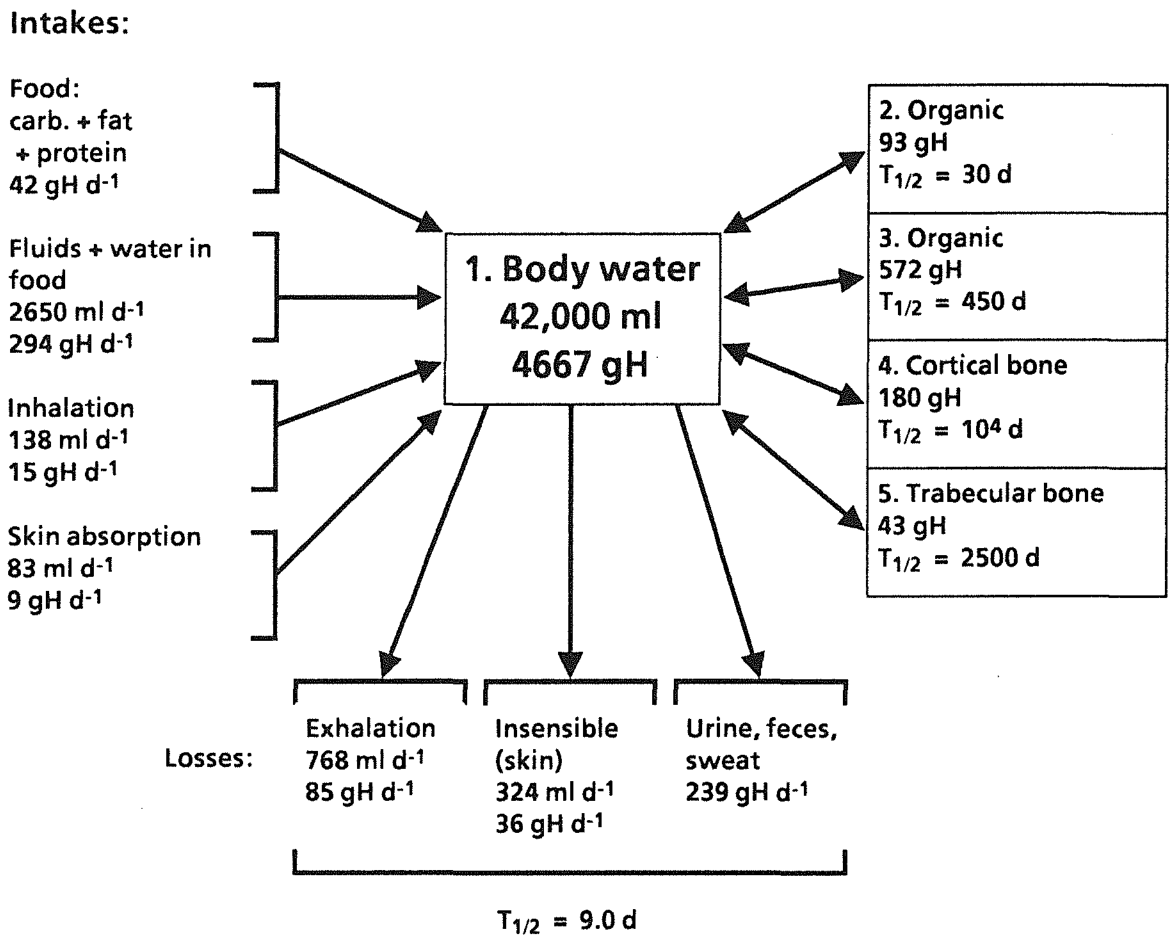

Figure 3.3: Compartment diagram for the dynamic and steady-state models of hydrogen uptake, retention, and removal according KILLOUGH (1982).

The organic compartments are based on the following assumptions:

The portion of dry organic matter of Reference Man (excluding fat, body water and mineral bone) is 9.5 kg assuming to contain 7 % hydrogen, or 655 g hydrogen. This portion is partitioned into two components to which he associates the longer-term removal half-times adopted by NCRP (1979) of T2 = 30 days and T3 = 450 days. The total daily exchange between these compartments and body water is assumed to be 3 g hydrogen, which is in agreement with the difference of the dietary intake and food oxidation of the Reference Man (ICRP 23, 1975).

On the basis of the cortical and trabecular bone turnover rates in Reference Man of 2.5 % and 10 % per year respectively and the respective hydrogen contents of 180 g and 43 g KILLOUGH calculates the transfer parameters for the bone compartments. The resulting removal half-times of 10,000 and 2,500 days, respectively, were not observable in existing experimental data. The model does not account for intake of tritiated organic compounds.

USSR model:
The two-compartment model utilized in the USSR for tritium dose calculations (BALONOV et al., 1984; BALONOV et al., 1974, Figure 3.4) is based on the proposal by SNYDER et al. (1968).

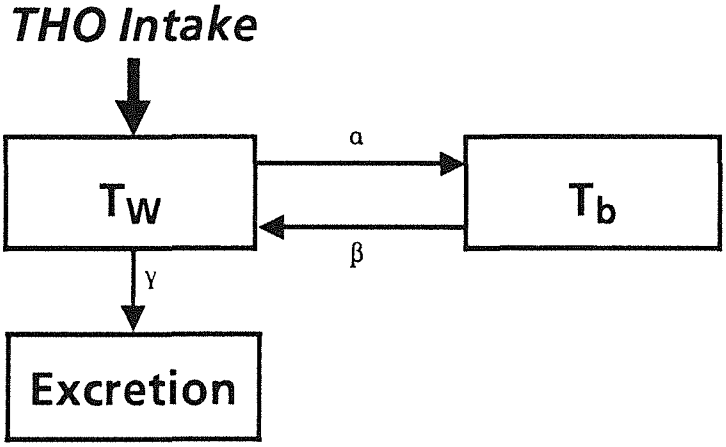

Figure 3.4: Model of exchange between H-3 in water (T_W) and that bound with organic structures (T_b) (SNYDER et al., 1968; BALONOV et al., 1974).

Intakes of tritiated water result in 100 % uptake into the free water compartment (T_W). For the tritium turnover in this compartment the physiological data of Reference Man (ICRP 23, 1975) have been adopted (10 days half-time in the 42 kg water compartment). Calculations based on this model showed that tissues with high water content, e.g. soft tissues excluding adipose tissue, are mainly irradiated from HTO. The contribution of bound tritium (T_b) make up about 4 %.

A main difference to other models is the use of a quality factor of 2 for tritium dose calculation.

4 Assessment of Existing Models

4.1 General Considerations

Following input parameters are necessary to estimate the dose to man due to the ingestion pathway after tritium releases to the atmosphere:

1. Tritium concentrations in drinking water.
2. Tritium concentrations in foodstuffs originating from contaminated vegetation.
3. Tritium concentrations in foodstuffs originating from animals which have incorporated tritium by inhalation, skin absorption and/or ingestion of tritiated drinking water and/or feed.
4. Average consumption rates of different foodstuffs.
5. The transfer rates of given foodstuffs.
6. The dose conversion factor.

Tritium concentration in drinking water and in vegetable foodstuffs can generally be estimated by dispersion models, which include appropriate submodels for soil and in plants.

The tritium concentrations of animal foodstuffs must be calculated by additionally compartment models. The pathways pasture grass to milk of lactating cows and to meat of beef are usually modelled by this way. Differences between models become obvious at this point, because some of them consider OBT and some not.

Different amounts of intake of a given foodstuff lead to different internal tritium concentrations. For the purpose of general assessment it is certainly not possible to consider the diet of each individual person, but estimates can be made for the mean rates of consumption as given in the Food Report (DGE, 1984). This report identifies the average consumption of individual foodstuffs by age groups and sexes. However, foodstuffs can origin from different areas which therefore may have different levels of contamination. Even, if the origin is the same, the contamination may vary during the course of a year.

To account for all variations in foodstuffs, calculations of dose would become too complex and difficult to be handled. Therefore, simplifications are necessary to enable for rapid decisions. For this reason, the foodstuffs were subdivided into several groups (for instance in BMI, 1979):

- milk and dairy products,
- meat and meat products,
- cereals and potatoes,
- vegetables and fruit,
- drinking water and beverages.

Most computer codes estimating the dose to man assume, that all food is produced in the contaminated area where the individual lives. The loss of the radionuclide from foodstuffs following harvest or slaughter, and prior to consumption (including processes like cooking) are neglected.

The uptake kinetics of tritium vary over time in weather conditions and growth. For the purpose of model calculations short term temporal variations are averaged over a period, e.g. the growing season.

A mean for calculation the dose is the ingestion dose factor which gives the amount of dose delivered per unit intake of a radionuclide. The dose factors are age-dependent and the unit is Sievert per Becquerel (Sv/Bq).

In the following consideration of different models only the submodels describing the ingestion pathway and their assumptions are compared. Table 4.1 lists these models and the countries and institutions applying them.

4.2 ISOLA IV

ISOLA IV is the Karlsruhe Nuclear Research Center code for calculation of the long term dose in the surroundings of nuclear facilities (HÜBSCHMANN and HONCU, 1986). The dose commitment of man due to chronic tritium exposure is estimated according the Guidelines under Section 45 of the German Radiation Protection Ordinance (BMI, 1979) from the ingestion of tritiated food.

Name of the code	Country	Institution applying the code
ISOLA IV	FRG	KfK (Kernforschungszentrum Karlsruhe GmbH)
TRITMOD	USA	SRL (Savannah River Laboratories)
OHTDC	Canada	CFFTP (Canadian Fusion Fuel Technology Project)
TRIDOS	UK	SRD of the UKAEA (Safety and Reliability Directorate of the United Kingdom Atomic Energy Authority)
COMA	UK	NRPB (National Radiological Protection Board)
UFOTRI	FRG	KfK
TRILOCOMO	Sweden	Studsvik

Table 4.1: Codes used to estimate the dose from atmospheric tritium releases.

HTO is the only tritium species considered. For its transfer from atmosphere to plant equilibrium conditions between the specific activities of tritium in the atmosphere at ground level and plant water are assumed (ANSPAUGH et al., 1973). The tritium concentrations in animal food products are estimated by the transfer factors feed / meat, feed / milk and feeding habits of animals. Beef was choosen as representative kind of meat. The dose to man is calculated from human consuming habits and the tritium dose factor.

This is a steady state model and applicable for chronic releases of tritium in the form of HTO.

4.3 TRITMOD

TRITMOD was developed at Savannah River Laboratories (SRL, US) by C. E. MURPHY Jr. (1986) for calculating the doses from the continuous release of either HT or HTO from a point source. It is a steady-state model which incorporates HT to HTO conversion and cycling of tritium in vegetation, dairy and beef cattle, and man. The tritium concentration in each compartment is calculated on the basis of a balance of tritium inputs and outputs. The code treats organically bound tritium separately from free-water tritium.

Tritium in vegetation:
The tritium content in the vegetation is devided into the water and the organic compartment. Uptake of tritiated water from the soil, diffusion of tritiated water between the plant leaf and the atmosphere, and the uptake and oxidation of tritiated hydrogen are taken into account. The uptake of HT and HTO via atmosphere is parameterized as deposition velocity.

The model assumes deposition velocities (v_d) for HT to vegetation of 5xE-4 m/s and for HTO to vegetation of 1xE-3 m/s. v_d is the reciprocal of the sum of the diffusion resistances of water vapour (atmospheric, boundary layer and vegetation resistance), corrected with the leaf area index. The different solubilities of HT and HTO in water are also considered in the formula.

Average values of diffusion resistances for Savannah River Plant vegetation were determined by studies on site (MURPHY et al., 1981, MURPHY, 1985, LORENZ and MURPHY, 1985).

The deposition velocities of HT to vegetation which were assumed in the model seem to be too high. They were used to get calculated tritium levels in the vegetation, that were similar to the measured values at SRL. These field data can be partially explained by the residual of higher tritium exposure during past years (MURPHY, personal communication).

The specific activity of the organic fraction in the vegetation is modelled as a steady state concentration. It is assumed to be in equilibrium with tissue free water tritium with an "accumulation factor" for tritium of 0.8 (a rejection) determined by GARLAND and AMEEN (1979). The fraction of nonexchangable tritium is estimated from experimental data (LUVALL and MURPHY, 1982). Tritium incorporation into the organic phase is assumed to come primary from tritiated water used in the photosynthetic processes and to a lower level from tritiated atmospheric hydrogen. While there is evidence that HT can enter directly organic matter, it is unclear what kind of process is involved (SWEET and MURPHY, 1984).

Tritium in cattle:
The water balance of the cattle is used to determine the tritium concentration in the water compartment (ANSPAUGH et al., 1973). Ingestion of water with food, water from oxidation of food, drinking water, water vapour exchange through breathing and diffusion through skin, and elimination in faeces and urine are taken into account. Slightly different values of some parameters are used for dairy and beef cattles. The distribution factors used to estimate the specific activity of the OBT are based on experimental data (VAN DEN HOEK, 1979).

Tritium in man:
The parameters of the human water balance are taken from ICRP 2 (1959). The assumptions are similar as for cattle but more complex because of the more complex diet of humans. The distribution coefficients for organic tritium retention by man are based on data for ingestion of tritiated food by cattle (VAN DEN HOEK et al., 1985) and rat (ARAI et al., 1985). A different distribution coefficient was derived for food high in proteins and fats, and for food high in carbohydrates. The division into these two types of food is based on information on the human diet (ICRP 23, 1975).

4.4 OHTDC

The Ontario Hydro Tritium Dispersion Code (OHTDC) is described in detail by BELL et al. (1985).OHTDC calculates tritium concentrations in air (dispersion model), soil and vegetation (terrestrial compartment model) and doses to man resulting from inhalation/immersion, and ingestion of food, milk, meat and water (dose model) from atmospheric releases of HT and HTO from a point source.

The code comprises two atmospheric dispersion models, one for acute and one for chronic releases. The code for chronic releases is based on the original TRITMOD code developed by MURPHY et al. (1979). The terrestrial compartment model and the dose model operate for both acute and chronic releases.

The movements in the terrestrial compartment model are described by transfer coefficients representing the mechanisms of dry deposition, rainfall, evaporation, transpiration, diffusion and mass transfer.

The tritium concentration in vegetation water is calculated from the HTO concentrations in air and in the rooting region of the soil using diffusion and transfer coefficients. HTO to OBT conversion in vegetation is not included in the model.

The calculation of the radiological dose to man resulting from the uptake of tritium is based partly upon the model recommended by the United States Nuclear Regulatory Commission in Regulatory Guide 1.109 (1977). The pathways of uptake are inhalation/immersion and ingestion of vegetation, meat, milk and water. The doses are calculated for a member of the critical group and average individuals in each terrain element.

The tritium concentration in drinking water is drawn from the deepest soil compartment and is assumed to have the same value.For calculating the tritium concentration in milk and the dose to man from milk ingestion the OHTDC cow-milk-model is used. It comprises one compartment, the cow's body water. Tritium uptake by the cow is calculated from inhalation of tritium in air and ingestion of tritium in vegetation and water. Drinking water is assumed to have the same concentration as the water in the topsoil compartment. The uptake and excretion rates for tritium are based on the water balance of the cow.

For calculating the tritium concentrations in beef animals and the dose to man from ingestion of beef, a similar model was constructed.

Sensitivity analysis have shown, that the calculated dose is very sensitive to the values of the vegetation compartment coefficients. The assumed water depth in soil corresponding to the equivalent rooting zone depends upon the type and maturity of vegetation. It was also found, that after an HT release the inhaled and ingested HTO contributes the major portion of the dose. HTO concentrations again depend mainly on the HT deposition velocity.

4.5 TRIDOS

TRIDOS was developed at the Safety and Reliability Directorate (SRD) of the United Kingdom Atomic Energy Authority (UKAEA) (BREARLEY, 1985). The model predictions are sensitive to the time within a year at which the accident occurs. That means for releases which occur near and before the end of the harvest season, the major pathway to man is assumed to be the consumption of food crops. For releases which occur after the end of the harvest and long before the beginning of the harvest the dairy and beef pathway dominates.

Vegetation activity is assumed to be proportional to the specific activity of the soil water, but the specific activity of atmospheric water vapour and its uptake by foliage is also taken into account. The formation of OBT is neglected. Cattle and sheep are assumed to eat pasture throughout the year. Man is assumed to obtain all his food from the contaminated area.

4.6 COMA

COMA (Compartment Model Analysis) was developed at the National Radiological Protection Board (NRPB, GB) (SHAW and HAYWOOD, 1986).

The description of the dynamic behaviour of the different forms of tritium in the food chains is possible adopting the so-called compartimental approach. The model consists of a series of inter-connected compartiments representing atmosphere, soil, plants and grazing animals. The rates of movement of tritium between the compartiments are expressed as transfer coefficients which represents a numerical solution of the problem. The time dependent behaviour of tritium can be described for transient and chronic release situations.

Tritium in vegetation:
The code ignores the transfer of HT from atmosphere to plants because the uptake is insignificant when compared to uptake from soil.

The transfer of HTO from atmosphere to plant water and organic substances is estimated assuming equilibrium conditions between the specific activities of tritium in atmosphere and plant water. For conditions where soil is uncontaminated the ratio is assumed to be 0.5 (ANSPAUGH et al., 1973). The transfer rate is calculated considering the water contents of different plant types (green vegetables, root crops, pasture and grain). For uptake of tritium into plant organic matter it has been assumed that the ratio between the specific activities of organic to aqueous components of plants is 1 at equilibrium. The specific activities are related to the content of hydrogen which is different in the organic and aqueous component.

This part of the model is not dynamic. For transient release situations, where equilibrium may not reached at all, the effect of this assumption is conservative.

For the loss of tritium from the plant the model assumes a half life of 1 hour for the loss of HTO by transpiration and 10 days for the loss of OBT to plant HTO.

Tritium in animals:
For modelling the transfer of tritium to animals the typical grazing animals of the UK, cow and sheep, were chosen. The routes of intake are inhalation, absorption through the skin and ingestion of pasture. The model assumes that animals continually graze a single area of pasture land and ingested intake is entirely in the form of grass. The consumption of drinking water is not included, because most animals derive their drinking water from local mains supply, which would almost certainly be uncontaminated. The incorporation of tritium into animal organic matter is included. But it is assumed that plant material ingested in the form of OBT is transferred to the HTO compartment of the animal due to the comparatively small amounts of plant OBT.

The model may be applied for acute and chronic exposures. In the case of constant atmospheric tritium concentrations the model predicts that equilibrium is reached after about 30 days. In a comparison with a steady state model (ANSPAUGH et al., 1973) it was shown that the levels of tritium concentration in vegetation and cows milk predicted by the NRPB model are slightly higher, because of the inclusion of OBT in the dynamic model.

Tritium in man:
The dose to man due to inhalation and ingestion of tritium is calculated using a committed dose equivalent per unit intake of $1.7 \cdot 10^{-11}$ Sv per Bq. It was assumed that drinking water is uncontaminated. Milk products are assumed to contain only the organic fraction of tritium in milk.

4.7 UFOTRI

UFOTRI (Unfallfolgenmodell für Tritium) presently under development at the Karlsruhe Nuclear Research Center (KfK) is a program for assessing the offsite consequences from accidental tritium releases (RASKOB, 1989). It will also be used as a submodule of the program system UFOMOD (Unfallfolgenmodell) (EHRHARDT et al., 1988) developed at KfK. In UFOTRI, the model describing the dispersion and reemission process (MOTTE = model for tritium transport in the environment) (RASKOB, 1988) is coupled to a modified version of the ingestion model in COMA (SHAW and HAYWOOD, 1986) describing the dynamic behaviour of tritium in the food-chains.

The process of tritium deposition on vegetation is described by a "resistance model", calculating the atmospheric, boundary layer and vegetation resistances to HTO deposition. The vegetation resistance is calculated according the mathematical model proposed by BELOT et al. (1979). To account for different species of vegetation, average values for day and night vegetation resistances (day: 4 s/cm, night: 60 s/cm) and for the leaf area index during the vegetation period from april to october (3 m^2/m^2) are assumed. These values are variable input parameters and can easily be changed by the program user. During rainfall in the daytime, the vegetation resistance is assumed to approach to zero. The high vegetation resistance during the night is assumed to remain constant in the case of raining.

The uptake of HTO by plant roots, which is dependent on the actual transpiration of the plant, is also considered in the model. It is assumed that 20 % of the transpired water, which is to replace by soil water, origin from the upper 5 cm and 80 % from deeper layers of the soil.

For the conversion of HTO to plant OBT an incorporation rate of 0.06 % per hour from the actual HTO concentration in tissue water in the daytime is assumed. The loss rate of OBT in the model is 0.3 % per hour from the actual tritium concentration in organic matter of plants.

By approximately 100 hours after the accidental event the parameters for the transfer between the compartments are recalculated every hour according the predominating weather conditions. Afterwards a modified COMA is started, which additionally includes the direct transfer of OBT from plants to animals and the transfer from OBT of plants and animals to OBT of milk.

UFOTRI can only be used for accidental releases of tritium to the environment.

4.8 TRILOCOMO

TRILOCOMO (EDLUND, 1988), a local compartment dispersion model for tritium in the form of HT and HTO, was developed at Studsvik (Sweden) and is based on H3DISP for atmospheric dispersion calculation and BIOPATH (BERGSTRÖM et al., 1982) for modelling the transport of radionuclides through the food chain to man.

The transfer of HT from the air to the vegetation is neglected due to its very low solubility in water. The uptake of HTO is calculated from the specific activity ratio between the tritium source, the atmospheric humidity, and the plant tissue water using the energy balance equation according MURPHY, 1984. The variations of the specific activity ratios with time and its dependence from air humidity, temperature, pressure and diffusion resistances are taken into account. The model is applicable for simulating short time effects concerning the tritium activity concentrations in air humidity, soil and vegetation water.

The BIOPATH submodel which is not tritium specific, predicts the long-term exposure of human population following radioactive releases using time dependent uptake coefficients in foodstuffs. The mathematical description of the dynamic behaviour of tritium in the different compartments was solved using coupled linear differential equations.

4.9 Comparison of the Models

The computer codes ISOLA IVand TRITMOD are able to predict the behaviour of tritium for chronic release situations. UFOTRI and TRILOCOMO can only be used for accidental releases and OHTDC, TRIDOS and COMA can calculate both release situations (Table 4.2). Submodels of each code can be of steady-state or of quasi-dynamic character.

	ISOLA IV	TRITMOD	OHTDC	TRIDOS	COMA	UFOTRI	TRILOCOMO
continuous release	x	x	x	x	x		
accidental release			x	x	x	x	x

Table 4.2: Comparison of the models regarding their applicability for chronic or accidental release situations.

The models presented in this chapter are now compared regarding their assumptions for tritium uptake by plants and its behaviour in animals and man.

Tritium in Plants (Table 4.3)

Assumptions	ISOLA IV	TRITMOD	OHTDC	TRIDOS	COMA	UFOTRI	TRILOCOMO
consideration of HT conversion to HTO and OBT	-	x	-	-	-	-	-
fixed specific activity ratios for uptake of HTO assuming equilibrium conditions between HTO in atmosphere and in vegetation	x 1.0				x 0.5		
calculation of the specific activity ratio considering the relative humidity and plant parameters		x	x	x		x	x
consideration of OBT formation	-	x		-	x	x	-

Table 4.3: Different assumptions in the submodels describing the behaviour of tritium in plants.

TRITMOD is the only model that considers HT deposition to vegetation and conversion to HTO and OBT. The other models neglect this pathway because it is thought to be insignificant for dose calculation. They calculate HT to HTO conversion in soil and subsequent uptake by plants. However, MURPHY (1986) demonstrated by comparison between simulations and measurements in the vicinity of the Savannah River Plant that the dose to man after chronic releases of HT may be higher, when direct HT conversion to OBT of plants is taken into account. Because of tritium transport through the food chain in the form of organically bound tritium and ingestion of tritiated compounds, this way reaches significance.

HTO uptake by plants from the atmosphere can be modelled assuming a fixed specific activity ratio between the tritium concentrations in air humidity and TFWT of plants (ANSPAUGH et al., 1973) or by calculating the specific activity ratio from atmospheric and plant parameters (e.g. resistance model). Fixed specific activity ratios are assumed in ISOLA IV (1,0) and COMA (0,5). TRITMOD, OHTDC, TRIDOS and UFOTRI calculate the specific activity ratio according the model proposed by BELOT et al. (1979) and take the uptake of HTO from soil also into account. In TRILOCOMO a more specific model is used, considering additionally the leaf temperatur in dependence of the radiant heat load (MURPHY, 1984).

The formation of OBT from HTO in vegetation water is only considered in TRITMOD, COMA and UFOTRI. In TRITMOD and COMA, the organic compartment in vegetation is assumed to be in equilibrium with TFWT, however, in TRITMOD, an isotopic fractionation factor of 0.8 for HTO incorporation into OBT by photosynthesis is considered additionally. In UFOTRI (code for accidental releases), the OBT compartment of vegetation is calculated by uptake from the actual TFWT concentration during the daytime (0.06 % per hour) and loss of OBT from the actual tritium concentration in organic matter (0,3 % per hour).

Tritium in Animals (Table 4.4)

Assumptions	ISOLA IV	TRITMOD	OHTDC	TRIDOS	COMA	UFOTRI	TRILOCOMO
representative animals chosen for the model:							
- dairy cow	x	x	x	x	x	x	x
- beef cattle		x		x			
- sheep				x	x		x
intake by ingestion of							
- drinking water		x	x	x	-	-	x
- pasture	x	x	x	x	x	x	x
intake by inhalation and skin absorption	x	x	x	x	x	x	x
considering of seasonal effects concerning feeding habits	x			x			x
considering uptake of OBT by ingestion		x			x	x	
considering of tritium incorporation into animal's OBT and into OBT of milk		x				x	

Table 4.4: Different assumptions in the submodels describing the behaviour of tritium in animals.

In all computer codes compared, the submodels describing the tritium behaviour in animals are based on the balance of tritium inputs and outputs using transfer coefficients to calculate the tritium transport between the different compartments. All models use dairy and beef cattle as reference animal. TRIDOS and COMA additionally consider tritium in sheeps, which are typical for agriculture in the UK.

Tritium intake by inhalation and skin absorption by animals is considered in all models, with the exeption of skin absorption by sheeps in COMA. The ingestion of tritium with drinking water by animals is neglected in COMA because it is assumed to be uncontaminated. OHTDC assumes for drinking water the same concentration as the water in the topsoil compartment.

All models consider the intake of pasture by grazing animals, but the grazing period is site specific. For instance in UK, the animals are assumed to graze all over the year, wheras in Canada, grazing is not practised during the winter.

Only TRITMOD, COMA and UFOTRI consider the ingestion of OBT by animals. In COMA it is assumed that OBT from plant material is only transfered to the water compartment of the animal. TRITMOD and UFOTRI also consider the transfer of plant OBT to animal's OBT (meat) and the transfer of plant and animal's OBT to milk.

Tritium in Man (Table 4.5)

Inhalation and skin absorption of tritium by man are calculated in all models. For the distribution of human diet average food habits for different countries, ages and sexes are assumed. In TRITMOD the human diet is divided in food rich in protein and fat and food rich in carbohydrate. Seasonal effects in the distribution of tritiated compounds in the human diet in dependence of the time of release are considered in TRIDOS.

The ingestion of tritium with drinking water by man is handled differently. OHTDC assumes that drinking water for man has the same tritium concentration as water in the deepest soil compartment considered. UFOTRI and COMA neglect the uptake of tritium by drinking water for accidental releases. The other models consider tritium uptake by drinking water but the way to estimate the tritium concentration is not described in the literature.

Assumptions	ISOLA IV	TRITMOD	OHTDC	TRIDOS	COMA	UFOTRI	TRILOCOMO
consideration of tritium uptake by inhalation and skin absorption	x	x	x	x	x	x	x
subdivision of the human diet into main groups	x		x	x	x	x	x
subdivision of the human diet in food high in proteins and fat and food high in carbohydrate		x					
consideration of tritium uptake by drinking water		x	x	x	-	-	
consideration of seasonal effects to the pathway of tritiated compounds				x			
dose conversion factor for adults(x 10^{-11} Sv/Bq)	1,6		2,0	1,7	1,7	1,7	1,7

Table 4.5: Different assumptions in the submodels describing the behaviour of tritium in man.

When the intake of tritium by the different pathways is estimated, the committed effective dose equivalent is calculated using the dose conversion factor for tritium in Sievert per Becquerel intake. This factor bases in all models on the data of water balance in the Reference Man according ICRP 23 (1975) and includes the quality factor, the biological half-life and the mass of affected tissue. All models use the quality factor of 1 and an average biological half-life of tritium in the body of 10 days. The dose conversion factors vary only slightly in the different models from 1,6 10^{-11} to 2,0 10^{-11} Sv/Bq for adults (whole body).

Only in TRITMOD the specific activity of the organic fraction in man is calculated additionally considering the uptake of food rich in protein and fat and food rich in carbohydrates. However, it is not clear in which way this concept will affect the resulting dose. The other models do not consider the increase of dose due to ingestion of OBT and incorporation of tritium into organic substances of man.

In the opinion of the authors, the submodels for tritium uptake by plants, animals and man in TRITMOD seem to be most realistic for the purpose to estimate doses due to chronic tritium releases, because modelling is combined with measurement studies at the Savannah River site over a long period of time. The model includes a number of parameters which are not considered in most other models, e.g. the transport of OBT from vegetation to animals and man.

Among the codes for accidental release situations, UFOTRI considers the most parameters relevant for the ingestion pathway. The code includes different stomatal resistances for day and night conditions and during rain, as well as OBT formation in plants and transfer of OBT to animal's food products.

5 Conclusions

This study was intended to collect the current knowledge on the behaviour of tritium in plants and in food chains after a release of HT or HTO to the atmosphere. For the purpose of dose estimations due to the ingestion pathway, the main parameters for tritium uptake and conversion processes in plants, animals and man were identified. A choice of existing computer codes has been compared regarding their assumptions to model these processes.

For routine releases, often rather simple models are used, as the so called "specific activity model", in which equilibrium conditions in the atmosphere-soil-plant system are assumed. Generally, this is a reasonable approach but relatively insensitive to changing activity concentrations and varying meteorological conditions. Furthermore, in case of HT emissions the reemission of HTO from the soil cannot be considered adequately.

Since equilibrium conditions cannot be anticipated for accidental releases, more sophisticated models are required for this purpose. They should be able to model the time dependent behaviour of tritium in the environment and its dependences from different environmental parameters in order to achieve an acceptable level of accuracy. Some models still assume steady-state character for certain submodels, which does not meat the real situation. Furthermore, most models neglect the influence of OBT intake what may lead to underestimation of the dose.

From the view of the authors, the parameter sets for the ingestion pathway of tritium, which are included in TRITMOD (SRL-code for chronic releases) and in UFOTRI (KfK-code for accidental releases), are recommendable for other model calculations (see also chapter 4). Variations of site specific data would be necessary for applications to another site. Some additional possibilities to refine the models are proposed in the following.

Plant models

1. Consideration of different plant species:
For modelling the tritium uptake by plants mean values for different species are assumed, for instance for stomatal resistances of leaves during day or night conditions and for the leaf area index, respectively. However, it is known that the uptake rates of atmospheric HTO are different for various plant species under the same environmental conditions. Since only leaves are considered, this would only be valid for grass and green vegetable, not for fruits and grain. Considering of different plant species is intended in UFOTRI.

2. Consideration of tritium translocation:
The transport of tritium from the leaves to other edible parts of different agricultural used plants is not considered in any model. This could be important for tritium releases occuring in the time before harvesting.

3. Consideration of diurnal variations in stomatal resistances:
Stomatal resistances, which directly affect HTO uptake from the atmosphere and also transpiration, can vary during the light period depending on the leaf water potential. These resistances will increase rapidly when the leaf water potential drops below a threshold value when plants are under water stress. The estimation of this effect would require informations of soil parameters.

For improvement of plant modelling, the knowledge on diurnal variations of stomatal resistances and photosynthetic intensity in dependence on photosynthetically active radiation, air temperature and humidity should be included. Observations of plant species, leaf area index and soil parameters should also be considered. These items are intended to be considered in UFOTRI. Since many parameters are site specific they should be included in the model as variable input parameter.

Animal models

All models contain the tritium cycling in dairy and beef cattle which is well studied. Depending on the consumption habits of man other representative animals are included, for instance sheeps for UK. Models could be refined by including the tritium uptake by other sources of meat like pig, fish and poultry. The fraction of OBT in meat and milk should be estimated, because it is essential for realistic dose estimations.

Human models

All models compared use the ICRP model based on human data for the ICRP Reference Man (ICRP 23, 1975) and assume a quality factor of 1 to calculate a dose conversion factor for tritium. But they neglect the increase of dose due to ingestion of OBT and tritium incorporation into human OBT. For dose estimations considering the ingestion pathway of tritium additionally to inhalation and skin absorption, it is necessary to take OBT into account, for instance by modified dose conversion factors for different kinds of food or by available models e.g. the ETNIER, TRAVIS, HATRIC model (ETNIER et al., 1984). The enhancement of the quality factor for tritium from 1 to 2, which was proposed by the Joint Task Group of the ICRP and the ICRU (ICRU 40, 1986), should also be taken into consideration.

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