

# Report Rapport



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Commission de contrôle  
de l'énergie atomique

ASSESSMENT OF THE SIGNIFICANCE  
OF ORGANICALLY-BOUND TRITIUM  
IN ENVIRONMENTAL MATERIALS

by

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ABSTRACT

The present state of knowledge of the significance, with respect to dose, of organically-bound tritium (OBT) in diet items has been reviewed. Ratios of the specific activity of the OBT to that of the free water (HTO) in foodstuffs have been commonly reported in the range of 1 to 4. A metabolism model of Etnier, Travis and Hetrick that takes direct assimilation of food OBT into account indicates that such levels result in a dose two to three times greater than that calculated solely on the basis of body water tritium content. Very high OBT/HTO values reported by Italian studies on food items are discounted. It is recommended that OBT/HTO measurements be done on Canadian diet items and that tritium metabolism models be more thoroughly evaluated.

RÉSUMÉ

Le présent rapport examine l'état des connaissances actuelles, du point de vue des doses, sur le tritium lié aux molécules organiques des aliments. On rapporte ordinairement que le ratio entre l'activité spécifique du tritium lié aux molécules organiques et de l'eau tritiée à l'état libre dans les aliments se situe aux environs de 1:4. Un modèle de métabolisme d'Etnier, Travis et Hetrick qui tient compte de l'assimilation directe du tritium lié aux molécules organiques dans les aliments, indique qu'un tel niveau entraîne des doses deux à trois fois supérieures à celles qui sont calculées uniquement à partir de la teneur en tritium dans les liquides organiques. Les ratios très élevés de tritium lié aux molécules organiques par rapport à l'eau tritiée qui ont été rapportés dans les études italiennes n'ont pas été retenus. Il est recommandé de mesurer les aliments canadiens pour déterminer le ratio du tritium lié aux molécules organiques par rapport à l'eau tritiée à l'état libre dans les aliments et d'évaluer plus en profondeur les modèles de métabolisme du tritium.

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SUMMARY

Reports of elevated levels of organically-bound tritium relative to aqueous tritium in foodstuffs and human tissue have raised concern about the effect of such conditions on the validity of dose estimates based on the assumption that all tritium intake is rapidly dispersed throughout the body water and may be monitored adequately by urinalysis. Exposure chamber and feeding studies have demonstrated that a portion of the organically-bound tritium (OBT) in diet items is assimilated quickly without oxidation and distribution in the body water. A transfer model incorporating a fast direct assimilation route for OBT in food indicates that dose estimated taking into account such assimilation will be significantly higher than that based simply on the HTO content of food and drink if the OBT/HTO ratio of the food is in the range 2 to 4, as has been observed commonly. Values much higher than this reported for diet items in Italy are discounted in view of the limitations of the sensitivity of the techniques used and lack of correlation with data from other laboratories. However, there are few other measurements of OBT/HTO in meats, the item that is particularly high in the Italian results. It seems advisable that some measurements of OBT/HTO be done on Canadian diet items to assess the situation here.

It must be pointed out that there should be no concern about underestimating the occupational dose of CANDU nuclear power station workers by limiting analyses to urine. Such occupational exposure is predominately to HTO and does not involve OBT which might by-pass the body water and be missed by urinalysis. Contact of bare skin with HT contaminated surfaces results in formation of OBT in the subject with the dose to skin at the point of contact being limiting. Doses from the OBT formed to other tissues are several orders of magnitude smaller. In any case where tritiated organic compounds are involved, examination of subjects for elevated OBT, e.g. in blood components, would be advisable.

In brief, OBT/HTO ratios greater than one have been commonly observed in foodstuffs and could result in a dose up to 3 times higher than that calculated solely on the basis of the HTO content of the food. Since normal concentrations of OBT in environmental materials are insignificant, this would only be of concern in the case of prolonged contamination of the general environment where foodstuffs are grown. However, in view of reports of very high OBT/HTO in meat, even though uncorroborated, it would seem advisable to have some reliable analyses done on Canadian diet items, particularly meats. There is no concern that monitoring of occupational exposure in CANDU power stations by urinalysis is inadequate.

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1.0 Forms of Tritium in Organic Matter

Tritium occurs in organic matter in three forms:

- i) In the free water associated with the matter.
- ii) Bound to oxygen, sulphur and nitrogen atoms in compounds of the material. This tritium is exchangeable with water in the system to varying degrees depending on molecular structures.
- iii) Bound to carbon atoms in the material. This tritium is non-exchangeable and only released on decomposition of the organic compound.

The exchangeable tritium in (ii) will be in equilibrium with the aqueous component of the system and so, from the point of view of specific activity, may be considered as part of the overall aqueous complement of the system. The free water tritium and the exchangeable tritium may have a different specific activity than the local environmental waters if the material has originated in a different region but will tend to equilibrate with local atmospheric moisture.

Component (iii) is designated Organically Bound Tritium (OBT). The OBT/HTO ratio of a material refers to the ratio of the specific activity (Bq/g hydrogen) of the carbon-bound hydrogen to that of the free water of the material. For ease of analysis exchangeable tritium is often measured along with carbon-bound tritium by simple removal of the free water (drying) and combustion in preparation for activity measurement. Carbon-bound tritium can be measured separately by equilibrating the material with tritium-free water prior to combustion. Carbon-bound tritium is sometimes designated Non-Exchangeable Tritium (NET) to differentiate it from combined exchangeable and carbon-bound tritium.

At the time of its formation, OBT has essentially the same specific activity as the water from which it was formed (OBT/HTO ratio = 1.0) since net isotopic fractionation in the biochemical reactions involved is slight.

## 2.0 Significance of OBT in the Diet

At present dose to the human body from intake of tritium is based on the assumption that all intake is in the form of water or is rapidly oxidized to water in the gut and is distributed uniformly throughout the body water (1,2). Bennett (3) has represented this by a three compartment transfer model, Figure 1, which consists of a body water reservoir,  $t_{1/2} = 10$  days, and two dependent organic hydrogen reservoirs,  $t_{1/2} = 30$  d and 450 d, corresponding to observed discharge rates from exposed individuals. Reports of elevated levels of organically-bound tritium (OBT) relative to aqueous tritium in plants and animals and evidence that the OBT content of animals fed tritiated food respond rapidly have raised concern that dose is underestimated in considering only intake to the body water compartment. Etnier, Travis and Hetrick (4) proposed a four compartment transfer model, Figure 2, to include a pathway for rapid assimilation of a portion of the OBT intake without oxidation. They argue that such a pathway is in line with known rapid assimilation of some lipid materials by the body. Furthermore, it has long been recognized that some amino acids are not synthesized in the body and must be acquired intact from the diet. Myers and Johnson (5) have reviewed the metabolism of organically bound tritium.

If OBT is assimilated directly it will not be evidenced in the body water. If the specific activity of intake OBT is the same as that of intake HTO, the dose estimate for chronic intakes will not be affected if the measured specific activity of body water is applied to the complete complement of H in the body and account is taken of the long residence time of OBT in the body. The Bennett model indicates a cumulative dose after 2000 d of 1.2 times that estimated from the free body water compartment alone. The four compartment model of Etnier et al yields a cumulative dose after 2000 d of 1.74 times the dose contributed by the free body water compartment alone. If the specific activity of intake OBT is greater than that of intake water, the dose estimate based on body water will be further underestimated. Figure 3 shows the dependence of dose estimate (cumulative after 2000 d) on the mean OBT/HTO ratio of the food items of the diet as indicated by the ET&H model. At a mean OBT/HTO ratio of 2 for the food items of the diet, dose estimate is 2.1 times that contributed by the free body water alone. It is evident that the OBT/HTO ratio of the diet should be taken into account in dose considerations.

## 3.0 OBT/HTO ratios in Environmental Materials

### 3.1 Measurement

Great care must be taken in the measurement of OBT at environmental levels. McFarlane (6) has pointed out the opportunities for cross-contamination of samples and contamination by exchange with the laboratory atmosphere. Most measurements have been done in laboratories at nuclear research centres where atmospheric HTO levels are usually elevated above the normal environmental levels one is trying to measure. For proper analysis the following points have to be achieved:

- a) A sample of at least 5 g is required to get enough counts for acceptable statistical accuracy. Commercial combustion units can accommodate a maximum of 1 g.
- b) Complete removal of free water from the material. The free water may have been contaminated by exposure to the lab atmosphere after sampling.
- c) Removal of exchangeable tritium from the material. This is accomplished by repeated soakings in T-free water or exchange with T-free steam.
- d) Final drying in the combustion system. This cannot be accomplished satisfactorily in a commercial combustion unit. The dried material is very hygroscopic and will pick up water from the lab air if exposed in transferring from a drying system to a combustion system. Table I illustrates contamination on drying samples in an air oven.
- e) Protection of all samples against exposure to the lab atmosphere before and after drying and combustion.
- f) Long counting time in a low background system.

Usually combustion in a quartz tube with a flow of dry O<sub>2</sub> is used to handle large enough samples. Strack (7) uses a plasma combustion system. A 2 litre Parr combustion bomb is used at Chalk River. It will handle up to 10 g fresh weight of material. In all cases drying in the combustion system is slow but necessary.

### 3.2 Data

Our first experience of OBT/HTO > 1.0 in plants growing under steady state conditions was with tomatoes and cucumbers grown in an experimental greenhouse near the Pickering Nuclear Generating Station (8). We observed OBT/HTO in the range 2.6 to 4.8 (Table II) in 18 samples. Since that time we have analyzed many samples of grass, leaves and pine needles around the CRNL site and found values from 0.6 to 4., but generally in the range 1 to 2. I believe the explanation in these cases is the following. Tritium in this area is dispersed as atmospheric HTO vapour and leaf moisture in equilibrium with this atmospheric vapour has a higher tritium content than the soil moisture which constitutes the main water body in the plant. Photosynthesis in the leaves produces organic matter having a higher tritium content than that in the main water component of the plant.

Table III presents data reported by Bogen et al (9) for New York soils, foods and human tissues. During the 1960's and 1970's when these samples were taken, there were wide fluctuations in the levels of HTO and HT in the environment as a result of thermonuclear bomb tests. It is easy to see how these fluctuations could put OBT/HTO in plants and animals out of equilibrium since the organic pool of H responds much more slowly than the aqueous pool for both intake and discharge. In Bogen's results, the high



values for soil must result from the persistence of OBT formed during the preceding decade when environmental levels were generally high. Workers in France (10), Germany (11) and Japan (12) have reported OBT/HTO ratios in the range of 1 to 4. Tables IV, V and Figure 4 present some of the data from the Karlsruhe Nuclear Research Centre. The values are variable, but consistently greater than 1.0. The Karlsruhe data for food items are particularly relevant as are those for Japan (Table VI).

Much higher values have been reported by Belloni and co-workers (13,14,15,16) for Italian food items (Table VII) and human tissues. The higher values for meat than for vegetation is inconsistent with experience on feeding OBT to animals, as discussed in the following section. Additionally they measured much higher concentrations of tritium in human blood than in urine (Table VIII) which in turn was higher than the local environmental HTO levels. They suggest that these levels result from high levels of OBT in the body and that the urine is not an adequate sample of the mean body tritium burden. They do not observe this high ratio of blood to urine tritium in the case of occupational exposure where the augmented exposure was not to OBT. Irlweck & Teherani (17) also measured high blood urine ratios but other workers (18) have not even at environmental levels (19). Consideration of the details of analysis of the Italian and Austrian workers raises doubts about the validity of their data. OBT was measured using a commercial combustion unit limited by sample size and cross-contamination opportunities. The difference in background reported for combustion samples and distilled urine sample is of the same size as the discrepancy between blood and urine counts. It is not evident why the counting background should differ for the two samples. The values they report for precipitation deviate significantly from data of the low level tritium laboratory of the IAEA (20,21) during the period this work was being done (Figure 5) suggesting a contamination problem in their laboratory or local environment.

In summary, it has been common experience to find OBT/HTO ratios  $> 1.0$  in vegetation and food items. Generally these have been around 2. Much higher values obtained in two laboratories are discounted on the basis of their analytical technique.

#### 4.0 Exposure Experiments

Numerous studies have been done on the incorporation of tritium into aquatic and terrestrial plants and animals. Many of these are included in the Bibliography and results have been compiled in Symposia Proceedings and Review Articles. A few will be discussed here to see if the phenomena observed may explain non-equilibrium OBT/HTO in plants and animals.

##### 4.1 HTO Exposure

Plants, trees, cows, pigs, mice, rabbits, fish, algae and man have all been studied.

The Tissue Free Water Tritium (TFWT) in plants, animals and fish quickly responds to HTO exposure, the rate depending on the size of the body water compartment of the specimen. The TFWT level usually rises to 60 - 80% of the tritium concentration of the exposure water, never exceeds it. In many experiments not all routes of water intake were tritiated, e.g. irrigation water but not atmospheric moisture, drinking water but not water contained in food or the atmosphere. The extent of approach to the concentration of exposure water was correspondingly affected. Aquatic plants and animals come closer to equilibrium than terrestrial plants and animals probably because of more complete tritiation of all intake routes.

Organic-bound tritium responds much more slowly to HTO exposure and seldom exceeds 20% of the concentration of the exposure water, often being only a few percent. Long-term approach to equilibrium is limited by remains of original tritium-free organic constituents in the specimen. On cessation of exposure, decrease of OBT is similarly slow, the OBT being decomposed and discharged as HTO.

Spencer (22) studied the varying uptake of tritium by the fruit, new foliage and old foliage of tomato plants exposed to HTO. Figure 6 shows the effect of exposure to atmospheric HTO on the OBT/HTO ratio of the different components. Old foliage takes up TFWT readily but synthesizes little OBT, so the OBT/HTO ratio is slow to rise. New foliage takes up TFWT quickly and also synthesizes OBT quickly, OBT/HTO approaches 1. However, with time OBT formation drops off and the OBT/HTO ratio falls. The green fruit has little exchange with atmospheric moisture, hence maintains a low TFWT. However, the fruit incorporates photosynthetic products translocated from the foliage, so the OBT exceeds the TFWT. With time more tritiated water comes into the fruit from the rest of the plant and the OBT/HTO ratio falls below 1.

#### 4.2 OBT Feeding

Fish, cows, pigs, mice, rats, rabbits have all been studied. The TFWT responds much more slowly and to much less extent than in the case of HTO administration. The TFWT remains lower than the OBT. On cessation of feeding, the TFWT decreases in parallel with the OBT.

The OBT of the specimen responds quickly and reaches higher levels than the TFWT, so  $OBT/HTO > 1.0$  occurs. The experience is similar with various animals and even fish. The OBT never exceeds the concentration of administered OBT. Hence, it does not seem that the OBT/HTO of meat could ever exceed that in feed.

#### 4.3 HTO and OBT Feeding

Figure 7 contrasts the effect of feeding a lactating cow tritiated water and tritiated fodder (23,24). Tritium appears in both the aqueous and the organic components of the milk in both cases, but with HTO administration the aqueous components show more rapid and much greater response than do the organic components. On the other hand, with tritiated

fodder, the organic components show the strong response and the aqueous components a weaker response. Here, an  $OBT/HTO > 1$ . is demonstrated in an animal product.

Rodgers (25) fed mice independently tritiated water, tritiated amino acids and both. His results on the rate of uptake and the rate of excretion after cessation of exposure are shown in Table IX. The steady-state specific activity of the TFWT was greater for HTO feed than diet feed and together they equalled that observed for simultaneous feeding of HTO and T-diet. Only a single reservoir was evident on uptake or elimination. The steady-state specific activity of the OBT was greater for diet feed than HTO feed. In fact, it appears to exceed the specific activity of the diet, but is less when the two are expressed per gram of protein, the tritium having been fed as tritiated amino acid. On elimination two reservoirs were evident, the first having a turnover rate  $1/2$  of that of incorporation and the second a turnover rate  $1/10$  that of incorporation. The rate of formation of HTO from T-diet feed was about the same as the rate of formation of OBT from HTO feed.

Moghissi, Bretthauer and Petzer (26) fed mice HTO and tritiated food of the same specific activity through three generations. Table X presents their results on the  $OBT/HTO$  ratio of various organs of all generations. They found no statistically significant deviation from a value of 1.0.

#### 4.4 HT Exposure

In some HT exposure studies, trace amounts of OBT were observed in pine needles (27), corn, tomato and poplar foliage (28) but the levels were so low as to be attributable to oxidation and uptake via the soil. In recent field experiments (29,30) no evidence for OBT formation directly from HT has been found.

Contact of bare skin with HT contaminated surfaces results in formation of OBT in the subject (18,31,32) with the dose to skin at the point of contact being limiting. Doses from the OBT formed to other tissues are several orders of magnitude smaller.

#### 5.0 Conclusion.

- i) It has been definitely demonstrated that OBT is directly assimilated to a limited extent in feeding experiments.
- ii) A proper transfer model for dose estimate should allow for this .
- iii) When  $OBT/HTO$  is  $> 1.0$ , dose will be underestimated if based solely on body water content.
- iv) Measurements by a number of workers indicate that  $OBT/HTO$  of food items is generally  $> 1.0$  but that it is unlikely that the mean  $OBT/HTO$  of diet will exceed 2.

- v) An OBT/HTO of 2.0 for the organic portion of food items results in a dose 2.1 times that contributed by the free body water alone according to the Etnier, Travis Hetrick transfer model.
- vi) Meat should never have OBT/HTO greater than that of the feed, according to results from experimental feeding.
- vii) The effect of OBT/HTO > 1.0 in environmental materials is important only in the case of long-term contamination of the area where foods and forage crops are grown. The immediate vicinity of a nuclear power station might be such a case.
- viii) Urinalysis is a valid monitoring technique for occupational exposure where no OBT is involved, such as in nuclear power stations.

## 6.0 Recommendations

It must be accepted that direct assimilation of OBT occurs and has an effect on dose. In any case where tritiated organic compounds are involved, examination of subjects for elevated OBT, e.g. in blood components, in hair, (33) would be advisable. In the case of exposed dial painters, no difference has been observed between blood and urine tritium levels.

The Etnier, Travis and Hetrick model, which gives the above estimates for the augmentation of dose due to elevated OBT/HTO ratios in food items, should be more thoroughly evaluated. It is more realistic than the NCRP model in representing recognized processes involved in the transfer of tritium from food to body tissue. However, it was "validated" by reference to the questionable data for Italian diet and tissue items (34). It should be validated against more reliable OBT/HTO data before it is accepted as taking proper account of the contribution of OBT to total dose.

Proper application of any "augmentation" factor to account for additional dose resulting from high OBT/HTO ratios requires knowledge of the OBT/HTO ratio of diet items. In view of the lack of data on the OBT/HTO ratio of Canadian foods, it would be worthwhile to have some reliable analyses done, perhaps on a site specific basis, to gain assurance that some unknown factor that may cause unusually high values such as reported for Italy is not present.

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**Subject Index for References**

**Reviews:** Ref. Nos. 5,24,37,42,48,51,61.

**Tritium in Man:** Ref. Nos. 2,3,5,13,15,17,18,19,31,32,44.

**Modelling:** Ref. Nos. 4,16,33,60,65,.

**Tritium in Environmental Materials:**

Ref. Nos. 7,8,9,10,11,12,14,18,20,21,36,40,45,52.

**Tritium in Terrestrial Plants:**

Ref. Nos. 6,7,8,10,11,12,22,24,27,28,29,30,37,46,47,49,53,59.

**Tritium in Aquatic Plants:**

Ref. Nos. 24,35,38,39,43,50,62.

**Tritium in Terrestrial Animals:**

Ref. Nos. 23,24,38,39,41,55,56,57,63,64,67,68,69,70,71.

**Tritium in Aquatic Animals:**

Ref. Nos. 24,34,38,39,41,43,50,54,58.

TABLE ICONTAMINATION of ORGANIC FRACTION  
on AIR OVEN DRYINGMeasured tritium conc'ns in Bq/L  
(combustion water or atmospheric moisture.)

Grass from CRNL Sites *****	Bomb Dried OBT ***** (Bq/L)	Oven Dried OBT ***** (Bq/L)	Atm. HTO during Drying ***** (Bq/L)
Emergency Basin	354	507	1343
Lower Bass Lake	118	935	-
Building 560	45	131	293

R.M. Brown Unpublished data.

TABLE IIOBT AND HTO in TOMATOES and CUCUMBERS GROWN  
near PICKERING NGS

## TOMATOES

\*\*\*\*\*

Harvest Date ***** (1979)	OBT (Comb. water) ***** (Bq/L)	Free Water T ***** (Bq/L)	OBT/HTO *****
Oct 22	1528 26	477	3.20
29	1458	540	2.70
Nov 05	1839	463	3.98
12	1624	444	3.66
19	951	340	2.79
26	614	222	2.77
Dec 03	1228	255	4.81
10	762	263	2.90
17	673	259	2.60

MEAN 3.27 0.74

## CUCUMBERS

\*\*\*\*\*

MEAN of 9 SAMPLES 3.25 1.76

OBT measurements by CRNL.

Free water T measurements by Ont Hydro.

Reference: Gorman D.J. "Levels of Tritium and Carbon-14 in  
Greenhouse Produce at Pickering NGS." Ont Hydro Report  
SSD-80-2 June 1980.

TABLE III

From: Bogen D.C., G.A. Welford, C.G. White. 1979. "Tritium Distribution in Man and his Environment." Proceedings IAEA Symposium on Behaviour of Tritium in the Environment. STI/PUB/498 pp 567-574.

## TRITIUM IN SOIL AND SEDIMENT

Location	Year	'Loose' tritium (pCi/l)	'Bound' tritium (pCi/l)	Ratio
<i>Soil</i>				
Chester, N.J.	1976	210 ± 40	2900 ± 100	13.8
	1977	200 ± 40	2650 ± 100	13.3
Lloyd, N.Y.	1976	190 ± 40	2800 ± 100	14.7
New York, N.Y.	1975	210 ± 40	2500 ± 100	11.9
	1976	180 ± 40	2600 ± 100	14.4
<i>Sediment</i>				
Sagamore Lake, N.Y.	1978	220 ± 40	3500 ± 400	15.9

## TRITIUM IN FOOD

Food type	'Loose' tritium	'Bound' tritium	Ratio 'Bound'/'Loose'
Milk	250 ± 40	460 ± 50	1.8
Bread	190 ± 40	800 ± 60	4.2
Lettuce	200 ± 40	680 ± 50	3.4
Root vegetable	190 ± 40	750 ± 50	3.9
Meat	180 ± 30	520 ± 40	2.9
Fish	160 ± 30	630 ± 50	3.9

## HUMAN TISSUE TRITIUM VALUES

Organ type	'Loose' tritium	'Bound' tritium	Ratio 'Bound'/'Loose'
<i>Subject 1</i>			
Lung	250 ± 30	530 ± 40	2.1
Liver	220 ± 30	480 ± 40	2.2
Kidney	260 ± 30	520 ± 40	2.0
<i>Subject 2</i>			
Lung	220 ± 30	420 ± 30	1.9
Liver	250 ± 30	400 ± 40	1.6
Kidney	190 ± 30	450 ± 40	2.4

TABLE IV

From: Strack S., L.A. Koenig. 1981. "Determination of Organically Bound Tritium in Environmental Samples by Application of the Oxidizing Plasma Technique." Kernforschungszentrum Karlsruhe Report KFK 3249.

Type of sample	Median of $R = \frac{OBT}{HTO}$
<b>Brussels Sprouts</b>  Heads Leaves Stems Roots	  2.4 2.3 1.5 1.4
Apples Pine Needles Beech Leaves Hornbeam Leaves	1.8 1.7 2.6 1.6

TABLE V

From: Koenig L.A., S.Fark, S.Wampelmann, K.G.Langguth, G.Pagliosa, D.Papadopoulos, S.Strack. 1987.

"Untersuchungen zum Transport von Tritium in der Umwelt." Table 3/8

Kernforschungszentrum Karlsruhe Report KfK 4131.

TRITIUM CONCENTRATIONS in PLANT FOODSTUFFS

from the 2nd DISTRICT (EGGESTEIN-LEOPOLDSHAFEN) in the vicinity of KfK

Plant Foodstuff	Sample Date (1984)	Water Content (%)	Tritium Spec. Act.		OBT/HTO Ratio	Sample Date (1985)	Water Content (%)	Tritium Spec. Act.		OBT/HTO Ratio
			HTO (mBq/g H)	OBT (mBq/g H)				HTO (mBq/g H)	OBT (mBq/g H)	
<b>Grains</b>										
Rye	09/08	11	9 ± 5	15 ± 5	1.7	08/08	12	6 ± 4	8 ± 5	1.4
Wheat	06/08	10	10 5	14 5	1.4	08/08	15	-	7 4	-
Corn	31/10	29	18 4	22 5	1.2					
<b>Leaf Crops</b>										
Leaf Lettuce	20/11	85	12 ± 5	19 ± 6	1.6	02/12	83	9 ± 5	12 ± 5	1.3
<b>Cabbages</b>										
White	04/07	93	8 ± 5	16 ± 5	2.0	08/08	93	5 ± 4	9 ± 5	1.7
Red	04/07	92	8 5	33 6	4.1	16/09	90	6 5	-	-
Savoy	04/07	93	8 5	8 5	1.0	16/09	91	6 4	7 4	1.2
"Kohlrabi"	04/07	92	9 5	32 6	3.6	26/06	92	-	8 5	-
Brussel Sprouts	20/11	83	13 5	14 5	1.1	02/12	85	17 5	13 5	0.8
<b>Root Crops</b>										
Carrots	04/07	89	7 ± 5	13 ± 5	1.9	26/06	91	-	14 ± 5	-
Radishes	04/07	95	6 5	18 5	3.0	08/08	95	8 ± 5	10 5	1.3
Beets	13/09	86	7 5	9 5	1.3	08/08	86	5 4	11 5	2.2
Potatoes	13/09	77	9 5	17 5	1.9	16/09	76	10 5	11 5	1.1
Asparagus	29/05	90	7 ± 5	28 ± 6	4.0	11/06	91	9 ± 5	12 ± 5	1.3
<b>Fruit</b>										
Strawberries	04/07	91	6 ± 5	22 ± 6	3.7	26/06	89	7 ± 4	6 ± 5	0.9
Apples	13/09	84	8 5	8 5	1.0	16/09	83	10 5	11 5	1.1
<u>Mean Values</u>			8 ± 5	17 ± 5	<u>2.16</u>			6.0 ± 4.4	11 ± 5	<u>1.33</u>

TABLE VI

From: Hisamatsu S., Y. Takizawa, T. Abe, T. Katsumata 1986. "Determination of Fallout Tritium in Food." Report of the Energy Project (Nuclear Fusion) of Ministry of Education and Culture of Japan. No. 60050024.

R1 Duplicate analysis of tritium in tissue binding water(Bq/l).

Sample	1st	2nd
Meats	3.3±0.3	3.4±0.3
Beans	4.3±0.3	4.1±0.3

R2 Variation of tritium concentration in tissue binding water(Bq/l).

Sample	1984	1985
Meats	3.2±0.3	3.3±0.3
Eggs	3.3±0.3	3.3±0.2

R3 Tritium concentration in food collected at Akita during 1985

Sample	Free water		Tissue binding Water		TB/FV(Bq/l)
	Bq/l	mBq/d	Bq/l	mBq/d	
Rice	4.4±0.3	100±6	3.2±0.3	350±30	0.73±0.07
Cereals without rice	2.5±0.3	90±9	3.6±0.3	130±10	1.4±0.2
Potatoes	2.4±0.3	110±13	2.7±0.3	24±3	1.1±0.2
Sugar	-	-	1.0±0.3	6.7±1.6	-
Confectionary	2.6±0.3	3.9±0.4	1.6±0.2	22±3	0.60±0.10
Beans	2.3±0.2	93±10	4.3±0.3	54±4	1.9±0.2
Soyasaues	1.3±0.3	24±5	5.3±0.2	16±1	6.0±0.8
Fruits	1.9±0.3	250±40	2.6±0.3	39±4	1.4±0.3
Green vegetables	2.4±0.3	110±10	2.6±0.3	4.9±0.5	1.1±0.2
Oter vegetables	1.2±0.2	230±40	3.0±0.3	26±2	2.4±0.5
Algae	-	-	1.2±0.2	2.4±0.3	-
Fish/Shellfish	(0.47±0.24)	(32±16)	1.9±0.2	29±3	(4.1±2.1)
Meats	2.7±0.2	120±10	3.3±0.3	46±4	1.2±0.2
Eggs	2.7±0.3	75±7	3.3±0.2	19±1	1.3±0.1
Milks	2.7±0.3	270±30	3.0±0.4	26±3	1.1±0.2
Oils	-	-	3.7±0.2	45±3	-
Total	2.0±0.1	1500±70	3.1±0.1	840±30	1.6±0.1

R4 Tritium concentration in tap water at Akita

Date	Bq/l
1984	
May	1.9±0.3
Jun	3.0±0.3
Aug	3.1±0.3
1985	
Feb	3.2±0.3
Mar	1.6±0.2
May	1.5±0.2
Jun	2.8±0.3
Jul	2.7±0.3
Aug	2.6±0.3
Sep	2.8±0.2
Oct	3.0±0.3

R5 Total Ingestion Intake of Fallout Tritium.

	Present Study				Bogen <sup>a)</sup>		Clemente <sup>b)</sup>	
	Water l/d	<sup>3</sup> H Bq/l	Bq/d	%	<sup>3</sup> H Bq/d	Bq/d	Bq/d	
Oxidation of Diet	0.27	3.1	0.84	14	7.0	36	} 15	
Free Water in Diet	0.77	2.0	1.5	24	9.9			
Other Water	1.46	2.6 <sup>c)</sup>	3.8	62				
Total		2.50 <sup>d)</sup>	6.1	100		51		

a): 1978, New York, Bogen, D.C. et.al, IAEA-SM-232/75.

b): 1975-1978, Italy, Clemente, G.F. et.al, IAEA-SM-237/64.

c): mean of monthly concentration in tap water during 1985, lacks of data were completed by linear completion.

d): (ratio of water intake to body weight for Reference man): (mean of body weight of reference Japanese man for both sexes).



TABLE VII

From: Tritium and plutonium content in the food-chain in Italy.  
 G.Ingrao, P.Belloni, G.F.Clemente, S.Di Pietro, G.Santori  
 Symposium on the Transfer of radioactive materials in the terrestrial  
 environment subsequent to an accidental release to the atmosphere.  
 Vol.II, page 481-495, Dublin, 11 April 1983.

**MEAN TRITIUM CONCENTRATION ± S.E. IN SINGLE FOOD ITEMS  
 AND TOTAL DIETS COLLECTED IN VARIOUS ITALIAN REGIONS.**

	BOND	HTO	OBT/HTO	
	Bq/g dry weight	Bq/ml		
MACARONI	75.8 ± 10.4	9.6 ± 2.6	14. +/-	4
MEAT	259.0 ± 96.2	15.9 ± 5.1	26. ±	13
FISH	≤55.5	23.7 ± 8.9	< 3.7	
CHEESE	≤55.5	45.9 ± 5.2	< 1.4	
FRESH MILK	86.2 ± 11.8	29.2 ± 7.4	3.6 ±	1
PROCESSED MILK	86.6 ± 16.3	9.3 ± 2.2	11.5 ±	4
ROOT VEGETABLE	≤55.5	14.4 ± 8.9	< 6.9	
LEAFY "	≤55.5	22.2 ± 5.2	< 4.5	
FRUITY "	65.9 ± 17.8	9.6 ± 2.2	12.3 ±	4
FRUIT	99.2 ± 25.5	27.7 ± 13.7	6.4 ±	4
EGG	≤55.5	12.2 ± 5.5	< 5.6	
WINE	---	19.6 ± 1.8	-	
DRINKABLE	---	9.6 ± 1.5	-	
TOTAL DIET	191.3 ± 70.3	50.3 ± 22.2	6.0	3.5

TABLE VIII

From: Belloni P., G.F. Clemente, S. Di Pietro, G. Ingraio. 1983. "Tritium Levels in Blood and Urine Samples of the Members of the Italian General Population and Some Exposed Subjects." Radiation Protection Dosimetry, Vol. 4, No. 2 pp 109-113.

### TRITIUM LEVELS IN BLOOD AND URINE SAMPLES

Tritium concentrations in urine and blood samples of members of the general population living in various Italian regions.

#### Group A (Castrovillari - CS)

Subject no.	Age	Sex	Body weight kg	Body height cm	Tritium concentration $\pm 2\sigma$		Ratio Blood/Urine
					Total blood pCi.ml <sup>-1</sup>	Urine pCi.ml <sup>-1</sup>	
1	37	F	95	172	6.7 $\pm$ 1.9	1.4 $\pm$ 0.5	4.78
2	20	M	52	158	5.0 $\pm$ 1.8	0.7 $\pm$ 0.5	7.14
3	38	M	73	165	13.4 $\pm$ 2.1	1.8 $\pm$ 0.5	7.24
4	37	F	50	160	5.8 $\pm$ 1.9	0.7 $\pm$ 0.5	8.28
5	40	F	82	173	12.7 $\pm$ 1.9	1.9 $\pm$ 0.5	6.68
6	38	M	68	162	—	$\leq$ 0.5	—
7	38	F	60	157	5.1 $\pm$ 1.8	0.9 $\pm$ 0.5	6.78
Average $\pm$ S.E.					8.28 $\pm$ 1.53	1.23 $\pm$ 0.22	6.82 $\pm$ 0.47

#### Group B (Novazza - BG)

Subject no.	Age	Sex	Body weight kg	Body height cm	Tritium concentration $\pm 2\sigma$		Ratio Blood/Urine
					Total blood pCi.ml <sup>-1</sup>	Urine pCi.ml <sup>-1</sup>	
8	43	F	50	175	4.1 $\pm$ 1.5	0.6 $\pm$ 0.2	7.42
9	29	F	58	170	4.0 $\pm$ 1.5	0.5 $\pm$ 0.2	7.69
10	24	F	90	180	5.4 $\pm$ 1.6	1.0 $\pm$ 0.2	5.31
11	38	F	53	155	4.3 $\pm$ 1.6	0.9 $\pm$ 0.2	4.89
12	37	F	70	170	4.0 $\pm$ 1.5	0.5 $\pm$ 0.2	7.44
13	44	F	70	163	7.2 $\pm$ 1.6	0.7 $\pm$ 0.2	10.30
14	26	M	75	170	7.5 $\pm$ 1.6	0.8 $\pm$ 0.2	9.98
15	21	M	70	170	5.1 $\pm$ 1.6	0.6 $\pm$ 0.2	9.05
16	47	M	73	180	6.5 $\pm$ 1.6	0.8 $\pm$ 0.2	8.15
17	25	M	63	167	10.4 $\pm$ 1.6	1.7 $\pm$ 0.2	6.07
18	36	M	75	168	2.7 $\pm$ 1.5	0.5 $\pm$ 0.2	4.98
Average $\pm$ S.E.					5.56 $\pm$ 0.66	0.78 $\pm$ 0.11	7.39 $\pm$ 0.58

#### Tritium concentration in urine and blood samples of some occupational exposed workers

Subject no.	Age	Sex	Body weight kg	Body height cm	Tritium concentration $\pm 2\sigma$		Sampling date month/year	Ratio Blood/Urine
					Total blood pCi.ml <sup>-1</sup>	Urine (HTO) pCi.ml <sup>-1</sup>		
1	56	M	83	169	200 $\pm$ 3	2200 $\pm$ 9	VI 1978	0.09
						122 $\pm$ 2	X	
2	56	M	50	154	141 $\pm$ 3	196 $\pm$ 3	VI 1978	0.72
						194 $\pm$ 3	X	
3	43	M	68	156	217 $\pm$ 3	244 $\pm$ 3	VI 1978	0.89
						211 $\pm$ 3	X	
4	41	F	90	164	188 $\pm$ 3	120 $\pm$ 2	VI 1978	1.57
						52 $\pm$ 2	X	

TABLE IX

From: Rodgers D.W., 1988. "Tritium Dynamics in Mice Exposed to Tritiated Water, Tritiated Diet or Both." Ontario Hydro Research Division Report 88-30-K.

**Estimated Steady State Activity and Rate Constants of Tissue Water Tritium of Mice Exposed to Tritiated Water, Diet or Both (Estimate ± SE)**

Tritium Source	Steady State Activity Q(eq) (kBq/g H)	Rate Constant	
		During Exposure k <sub>1</sub> (/d)	Following Exposure k <sub>2</sub> (/d)
Water (22.9 kBq/g H)	15.9 ± 0.1	0.68 ± 0.03	0.46 ± 0.02
Diet (34.4 kBq/g H)	7.0 ± 0.1	0.34 ± 0.03	0.27 ± 0.02
Both	22.8 ± 0.1	0.55 ± 0.02	0.41 ± 0.01

**Estimated Steady State Activity and Rate Constants of Organically Bound Tritium of Mice Exposed to Tritiated Water, Diet or Both (Estimate ± SE)**

Tritium Source	Compartment Activity			Rate Constant		
	During Exposure	Following Exposure		During Exposure	Following Exposure	
	Q(eq) (kBq/g H)	Q <sub>(s)</sub>	Q <sub>(s)</sub>	k <sub>1</sub>	k <sub>2</sub>	k <sub>3</sub>
Water (22.9 kBq/g H)	3.5 ±0.1	1.6 ±0.5	2.7 ±0.6	0.16 ±0.01	0.31 ±0.23	0.029 ±0.001
Diet (34.4 kBq/g H)	42.6 ±0.8	20.3 ±8.5	27.8 ±9.6	0.11 ±0.01	0.21 ±0.15	0.026 ±0.013
Both	46.1 ±0.8	26.1 ±4.0	29.7 ±4.2	0.11 ±0.01	0.36 ±0.14	0.023 ±0.006

TABLE X

From: Moghissi A.A., E.W. Bretthauer, R.G. Patzer. 1987. "Biological Concentration of  $^3\text{H}$ ." Health Physics Vol. 53 No. 4 pp 385-388.

Average ratio of organic to aqueous  $^3\text{H}$  (OT/OA) in various organs of rabbits.

Tissue	P	F <sub>1</sub>	F <sub>2</sub>	F <sub>3</sub>	Average
Liver	1.17 ± 0.20	1.05 ± 0.12	0.933 ± 0.202	0.938 ± 0.093	1.02 ± 0.08
Kidney	1.08 ± 0.11	1.06 ± 0.15	0.926 ± 0.043	0.960 ± 0.113	1.01 ± 0.06
Lung	0.994 ± 0.130	0.950 ± 0.246	0.990 ± 0.086	0.945 ± 0.083	0.97 ± 0.08
Brain	0.885 ± 0.052	0.901 ± 0.110	0.711 ± 0.057	0.904 ± 0.023	0.85 ± 0.05
Muscle	0.969 ± 0.097	1.05 ± 0.19	1.04 ± 0.08	1.13 ± 0.08	1.05 ± 0.06
Skin	0.963 ± 0.189	0.967 ± 0.123	1.08 ± 0.10	1.00 ± 0.12	1.00 ± 0.07
Average	1.01 ± 0.07	1.00 ± 0.08	0.95 ± 0.05	0.98 ± 0.04	0.98 ± 0.03

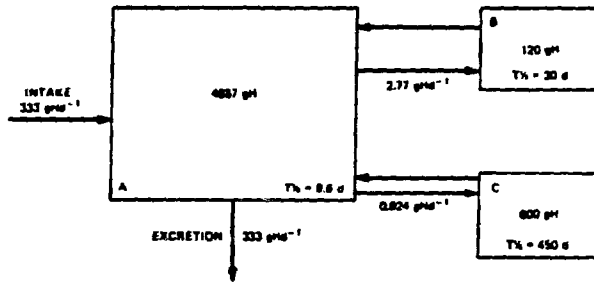


Figure 1: National Council of Radiation Protection and Measurements three-compartment model of hydrogen in the body. Reference (2).

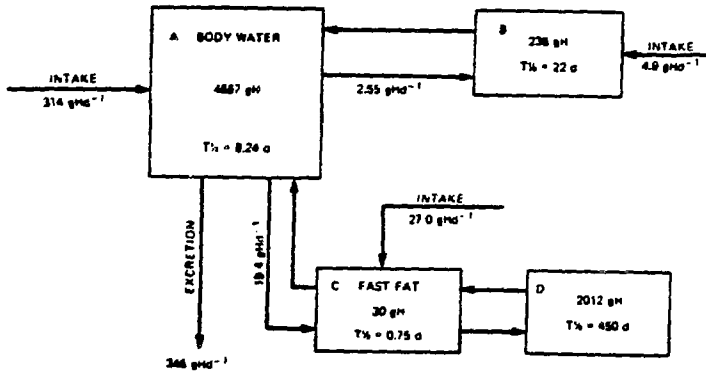


Figure 2: Four-compartment model of hydrogen metabolism of Etnier, Travis and Hetrick. Reference (4).

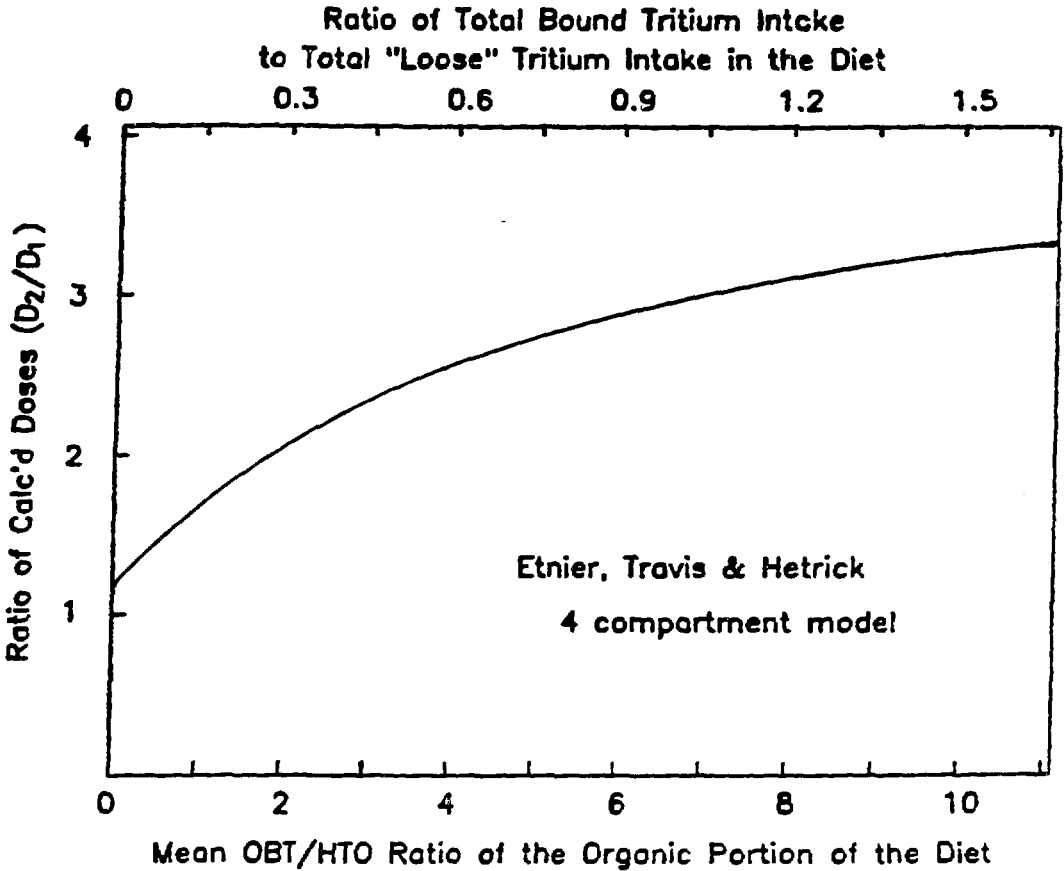


Figure 3: Etnier, Travis & Hetrick metabolism model prediction of the ratio of cumulative total body dose,  $D_2$ , to free body water dose,  $D_1$ , after 2000 days as a function of the ratio of OBT to HTO in the diet. Adapted from Fig.4 in Reference 3.

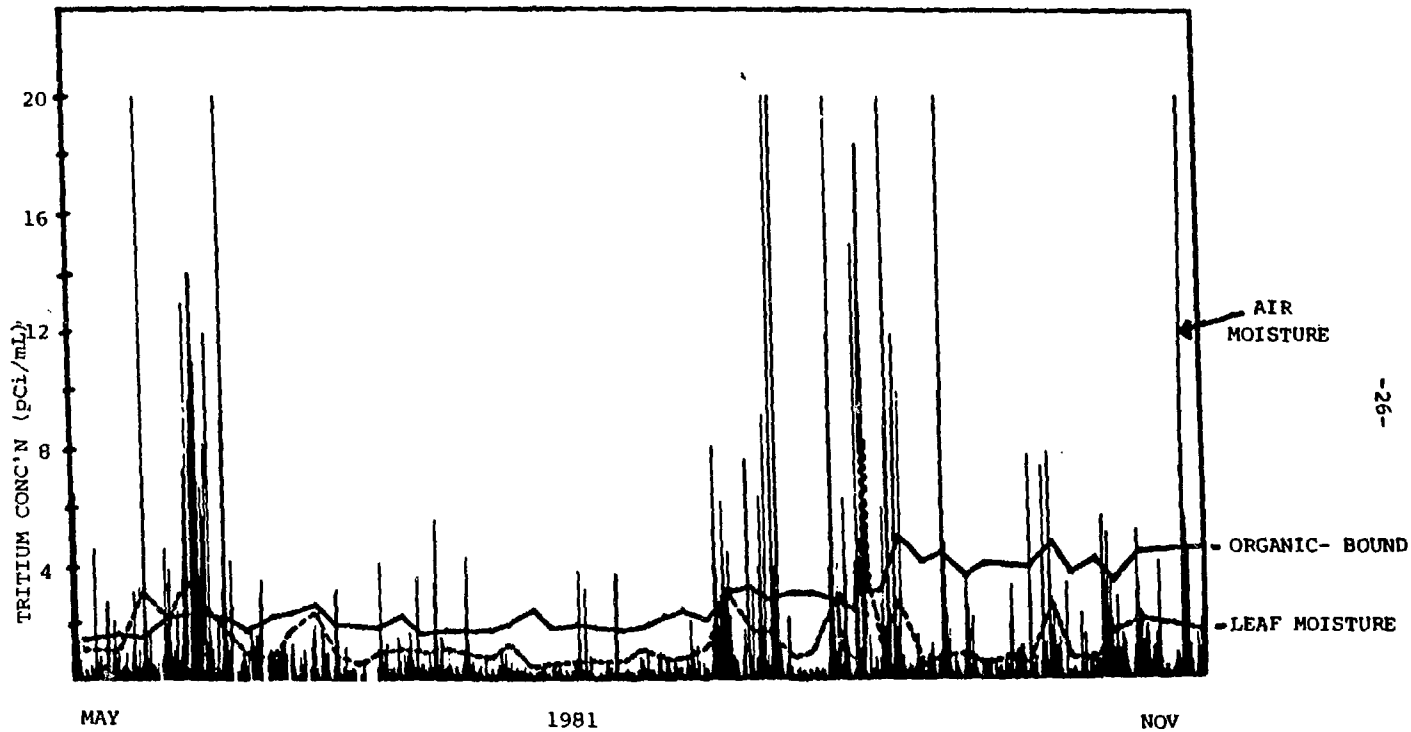
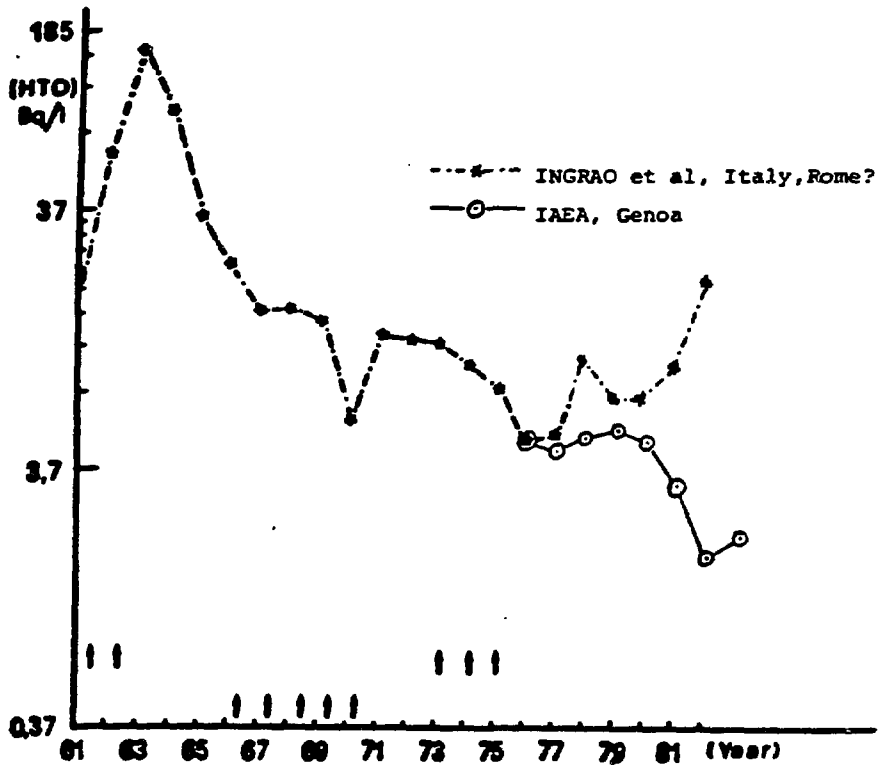


Figure 4: Tritium in Beech Leaves and Air Moisture at Karlsruhe, West Germany.

S. Strack, Annales de l'Assoc. Belge de Radioprotection. Vol. 7 No. 3,4 (1982).



TRITIUM IN PRECIPITATION IN ITALY

Figure 5:

From: Tritium and plutonium content in the food-chain in Italy.

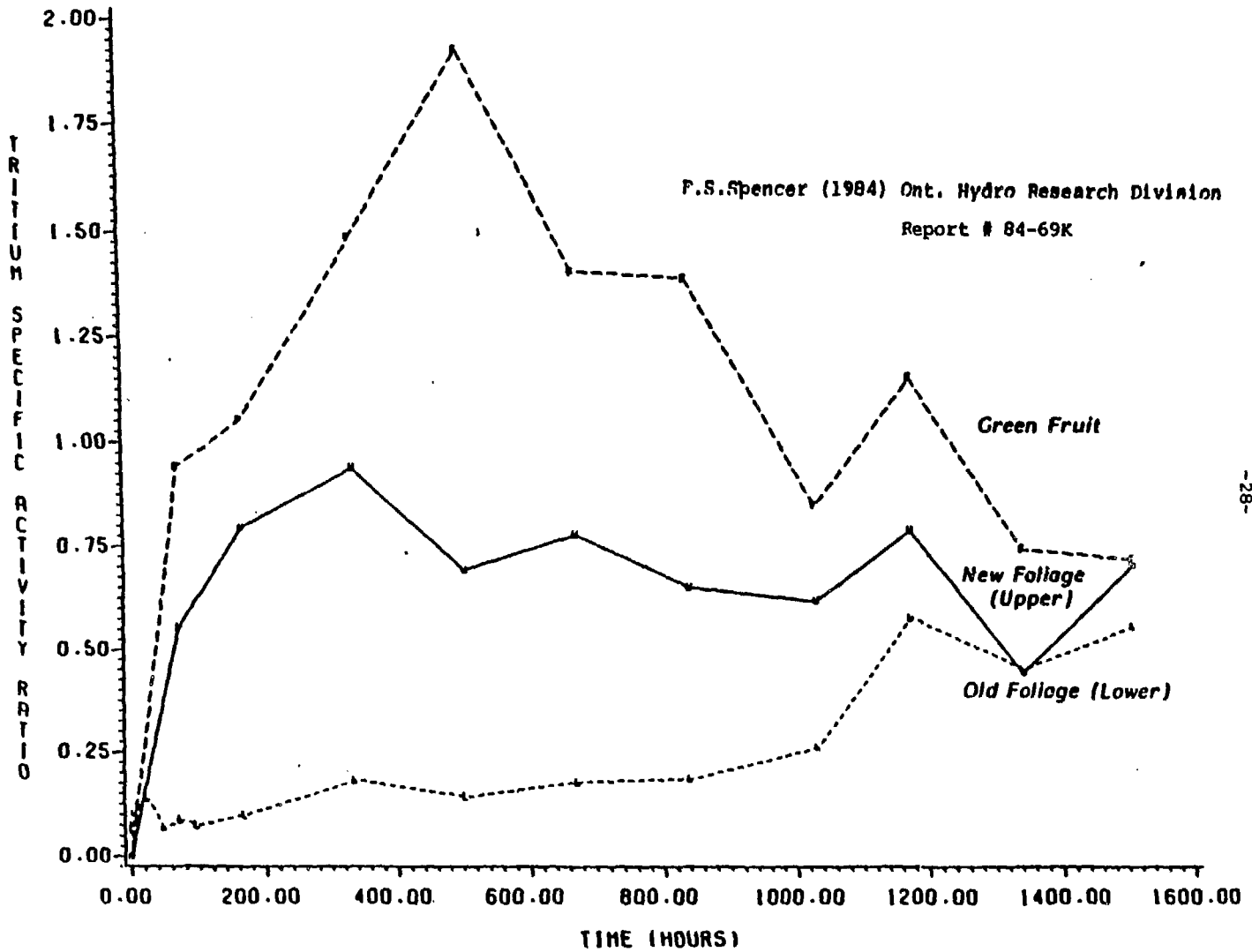
G.Ingrao, P.Belloni, G.F.Clemente, S.Di Pietro, G.Santori

Symposium on the Transfer of radioactive materials in the terrestrial environment subsequent to an accidental release to the atmosphere.

Vol.II, page 481-495, Dublin, 11 April 1983.



F.S.Spencer (1984) Ont. Hydro Research Division  
Report # 84-69K



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FIGURE 6  
CHANGES IN TRITIUM SPECIFIC ACTIVITY RATIO  
OF OAT TOLERANT DURING H<sub>2</sub>O UPTAKE

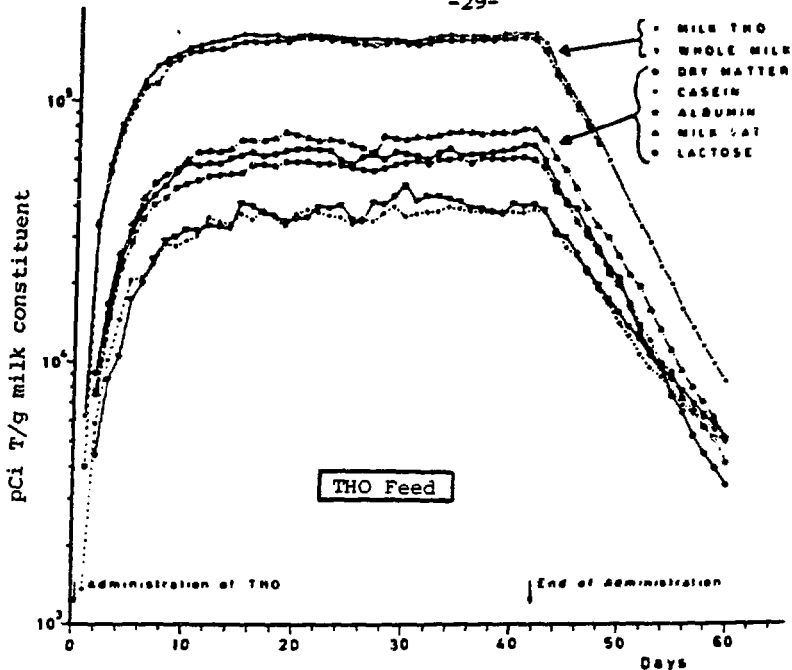


Figure 7a: The evolution of <sup>3</sup>H activity in milk and in milk constituents after continuous administration of THO for 41 days to a lactating cow. From: Potter G.D., Vattuone G.M., McIntyre D.R. Health Physics 22 (1971) 405.

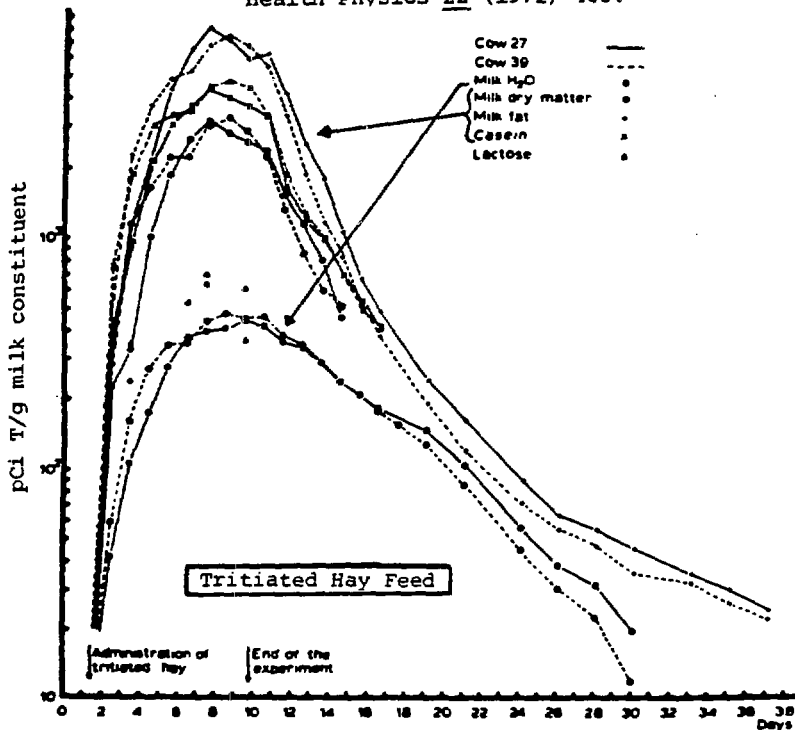


Figure 7b: Tritium activity in various milk fractions (milk H<sub>2</sub>O, total dry matter, milk fat, casein and lactose) after administration of organically bound tritium (tritiated hay) to two lactating cows. From: Fig.5 of IAEA Tech. Report No. 207 (1981).