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Investigation into Tritium Behaviour in Chinese cabbage and Rice after a Short-term Exposure of HTO

Sang-Bog Kim, Myung-Ho Lee, Gun-Sik Choi,
Young-Ho Choi and Chang-Woo Lee
Korea Atomic Energy Research Institute

HTO 피폭후 배추와 벼에서 삼중수소 거동

김상복 · 이명호 · 최근식 · 최용호 · 이창우
한국원자력연구소

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Abstract - Tritium concentration in the atmosphere during HTO exposure was different between two experiments due to different velocity of HTO evaporation. Assimilation rate of the rice plant was considered to be higher than that of Chinese cabbage. The uptake of atmospheric HTO into tissue free water tritium(TFWT) was higher in rice plants than in Chinese cabbage. However, organically bound tritium(OBT) concentration is relatively higher in Chinese cabbage than in rice. The specific activity ratio(SAR) increased slowly after HTO exposure and decreased gradually with time. The behaviour of HTO in the soil was affected by the environmental conditions.

요약 - HTO로 피폭시키는 동안 피폭상자내 공기중의 삼중수소 농도는 트리튬 용액의 증발속도 뿐 아니라 주변환경의 영향으로 피폭시기마다 다르게 나타났다. 어린 벼의 식물 대사율은 배추의 대사율보다 다소 높게 측정되었다. 증발된 HTO가 식물의 조직자유수에 흡수되는 정도는 배추보다 벼에서 높게 나타났다. 그러나 조직결합수의 농도는 어린 벼보다 배추에서 오히려 높게 계측되었으며 HTO 피폭 후에 조직자유수와 조직결합수의 비율은 처음에 증가하다가 시간에 따라 천천히 감소하였다. 오염된 토양에서 자라난 새로운 식물은 대부분 1보다 큰 SAR 값을 나타내었다. 피폭 약 1달 후 토양의 깊이에 따른 트리튬의 농도는 기상환경에 의해 큰 영향을 받았다.

INTRODUCTION

Tritium plays an important role in safety considerations for nuclear facilities. It is released from nuclear facilities during normal operation as well as at the time of an accident mainly in the form of tritiated hydrogen (HT) or tritiated water (HTO). HTO is the most abundant form of tritium and has been estimated to be 25,000 times more radiotoxic to human-being than HT [1,2]. For this reason, an understanding of the behaviour and fate of HTO in the crops is of particular importance and it is necessary to know the subsequent fate of tritium as non-exchangeable organically bound tritium(OBT) within crops. OBT has longer residence time in organisms than tritiated water [3]. In addition to tissue free water tritium(TFWT), the significance of OBT with respect to the dose has been recognized recently [4,5].

HTO is easily absorbed by plant leaves from soil and air, but is also easily removed from them by leaching with tritium free water or drying up. On the contrary, tritiated water is not so easily absorbed by fruits and grains, but once taken up by them, it is not easily removed. The ingestion dose due to an accidental or incidental short-term tritium release can be dominated by the consumption of wheat and rice, if tritium is incorporated into leaves and accumulated in grains during the grain-filling period until harvest. The 'Plant-OBT' model, developed

in FZK(Germany) should be adapted and applied to rice. Chinese cabbage which are the most important crop plants for nutrition in the oriental countries [6,7].

This experiment was carried out to evaluate the consequences of short-term tritium release to the atmosphere on tritium incorporation into crop plants and to compare uptake and formation of OBT in rice and Chinese cabbage.

EXPERIMENTAL METHOD

Plant material

A japonica type of rice (*Oryza sativa L.*) were sown in the pot (60x50x15cm) after sterilization using a properly diluted pesticide solution. Rice seedlings were transplanted into other pots(dia 25x20cm) and they were provided with water as required. The rice plant did not so well develope due to low temperature and insufficient to sunlight. Unfortunately rice plant could not grow to produce grains. Rice plants were exposured to HTO at their heights of about 25cm. Chinese cabbage(*Brassica chinensis* var. Hong Kong, F1-Hybride) was cultivated in the same type of pots containing agricultural soil. Chinese cabbage is usually cultivated very different soil compared to rice field.

Gas exchange measurement

Simultaneous measurement of net photosynthesis and transpiration were made with an open circuit gas exchange system consisting of a temperature and humidity controlled measuring cuvette and a controlling unit(Walz Meβ- und Regeltechnik, Effeltrich, FRG). A part of the leaf(4-5㎠) was enclosed in the transparent cuvette(volume about 1 liter) and CO_2 and H_2O concentration differences between the airs entering and leaving the cuvette were measured with high sensitive infrared gas analyzers (BINOS, Leybold-Heraeus, Hanau, FRG), averaged over periods of 5 min.

HTO exposure

The potted plants have been enclosed in a plexiglas box made of three parts: 2 side parts and one cover(ground area 30x30 cm, height 100 cm). Inside was a fan to prevent gradient build up and to minimize the boundary layer resistance of leaves, sensors for temperature and relative humidity. Air was drawn out for measurement of tritium concentration by the bubbler method(0.3 l/min) and by the tritium monitor (0.5 l/min). 2 l/min were drawn out to measure absolute CO_2 concentration and to pass the cuvette of the gas exchange measuring device in order to measure the rate of photosynthesis and transpiration of the leaf not exposed tritium but at the same temperature and light condition than the exposed plants. This gas stream was passed through a cooling trap in dry ice to prevent interaction of water vapour with CO_2 measurement. The water of this cooling trap was measured and compared with tritium measurements in atmosphere. Five minutes before start of exposure, the box was closed by the cover. HTO vapour was generated in the box by continuously evaporating 10 MBq HTO in 2 ml for about 50 min by the heating unit. One hour after start of exposure, the cover was opened and an additional fan was turned on to flush the box with fresh air for one minute. Then one side part was removed and plant and soil samples were taken.

Analytical methods

The tissue water was extracted by freeze drying the complete plant and soil samples and measured by liquid scintillation counter (1414 WinSpectral, Wallac). The dried plant samples were pulverized and exchangeable bound tritium was removed by wetting the dried samples with 15 ml tritium free water and freeze drying again. This procedure was repeated when the tritium concentration of the first exchange water was above 50 Bq/ml. After this treatment the dry samples were exposed to a stream of wet air(2 liters/min) in a glass vessel containing about 20 samples spread on petri dishes. The air leaving the vessel was cooled to trap air humidity which was used to measure the degree of exchange. The treatment was stopped when the tritium concentration in the condensation water was below 0.1 Bq/ml (about 5-10 days after beginning of the exchange). Before combustion with the oxidizer 306 (Canberra Packard), the samples were dried over P_2O_5 (Sicapent, Merck, FRG) in vacuum for two days and pressed to pellets of 0.5g at maximum. OBT was determined as specific activity in Bq per g dry matter. For comparison with the TFWT, the

Table 1. Meterological data and tritium concentrations inside the box during 1 hour exposure.

Parameter	Rice experiment	Chinese cabbage experiment
Mean temperature (°C)	19	24
Mean relative humidity(%)	82	66
Mean PPFD (μ mol m^{-2} s^{-1})	270	740
Mean HTO concentration in the atmosphere		
- Bq m^{-3} of air	4.7 x 10^6	5.1 x 10^6
- Bq ml^{-1} of air humidity	380 x 10^3	444 x 10^3
Integrated HTO concentration in the atmosphere (Bqs m^{-3})	1.85 x 10^{10}	1.85 x 10^{10}

PPFD : Photosynthetic Photon Flux Density (1 μ mol m^{-2} s^{-1})

specific OBT concentration was converted into the unit Bq per ml of oxidation water referring to the assumption that 1 g of dry matter yields 0.6 ml oxidation water.

RESULTS AND DISCUSSION

Exposure conditions

Tritium concentration in the atmosphere during the 1 hour's exposure was different between the two experiments because of differences in the velocity of HTO evaporation. Table 1 shows meteorological data and tritium concentrations inside the exposure box during HTO exposure. During the experiment, HTO concentration inside of exposure box was controlled by electrical heating and volume of tritium source. But tritium concentration in air was depend on external environmental condition such as temperature, humidity etc. The data shows variation between rice exposure and cabbage exposure. During the rice experiment, HTO concentration of exposure box was slightly higher than during the cabbage experiment but maximum value of HTO concentration was appeared in the cabbage experiment(Fig. 1,2). It means that it is very difficult to have a constant exposure condition in the field experiment.

Relationship between sunlight and assimilation

In addition to growing condition, the shape of leaves was related to the basic metabolism of each plant. Rice plant has narrow and long leaves but cabbage has very big and broad ones which may be expected to maximize the assimilation [8]. The light intensity(Fig. 3,4) is given as measured in Photosynthetic Photon Flux Density(PPFD). The assimilation rate of rice plant was saturated at about 600 PPFD where it was about 8 μ mol/m^2s. In case of Chinese cabbage, there was also a significant relationship between assimilation rate and light intensity. The degree of stomatal opening mainly depends on light intensity. Most plants close their stomata during the night. Stomata can also be closed during the day when plants are stressed by high atmospheric demand and dry soil [9].

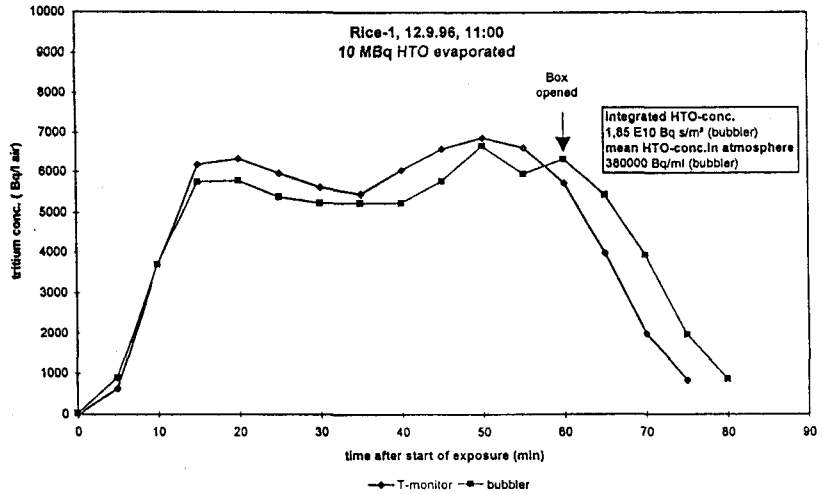

Fig. 1. Measured HTO concentration in the exposure box during the HTO evaporation at the rice experiment.

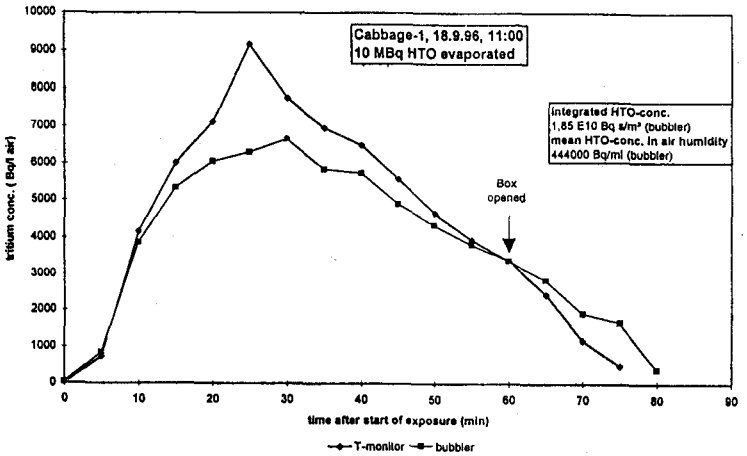

Fig. 2. Measured HTO concentration in the exposure box during the HTO evaporation at the Chinese cabbage experiment.

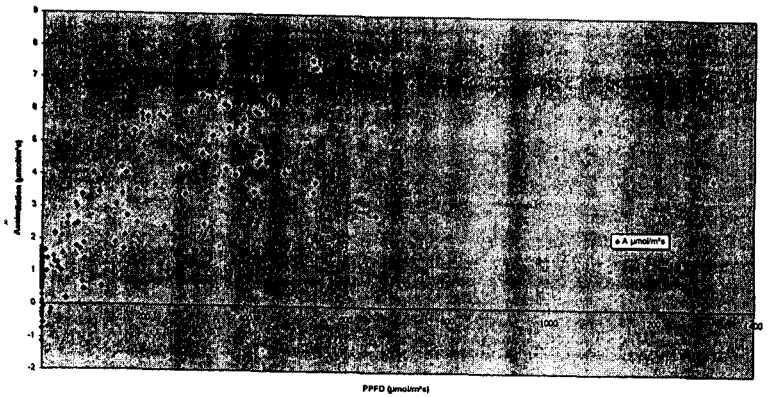

Fig. 3. The relationship between light intensity and metabolic assimilation in the rice plant.

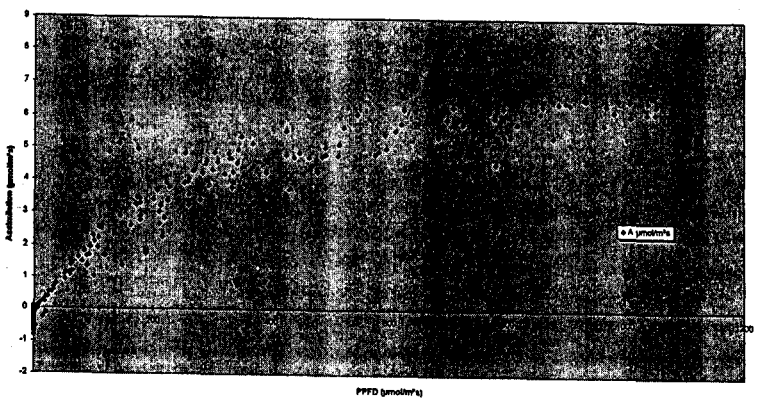

Fig. 4. The relationship between light intensity and metabolic assimilation in the Chinese cabbage.

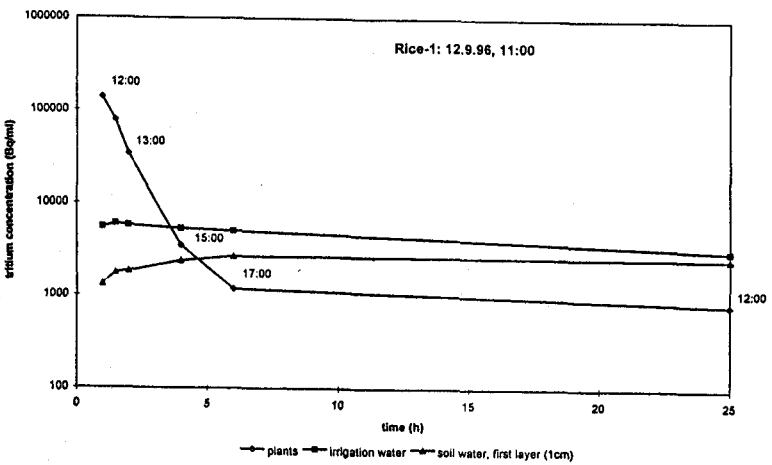

Fig. 5. Tissue free water tritium concentrations of rice plant and soil water at different times after HTO exposure.

Table 2. Uptake of atmospheric HTO into TFWT, irrigation water and soil water in relation to the mean HTO concentration in the air humidity and loss of TFWT one hour and one day after HTO exposure.

Parameter	Rice experiment	Chinese cabbage experiment
TWT concentration in plants relative to the mean HTO concentration in the air humidity	35 %	24 %
HTO concentration in the irrigation water of rice plants relative to the mean HTO concentration in the air humidity	1.4 %	-
HTO concentration in the first layer of soil(1cm) relative to the mean HTO concentration in the air humidity	0.3 %	14 %
Loss of TWT concentration in plants one hour after HTO exposure	76 %	54 %
Loss of TWT concentration in plants one day after HTO exposure	99 %	99 %

For modelling the plant growth, the rate of photosynthesis has to be considered responsible for the accumulation of *in vivo* synthesized organic material based on the assimilation of plant. For plants like rice and Chinese cabbage, the typical curve of the rate of photo- synthesis versus PPFD rises steeply until a plateau is reached, i.e. further increase in PPFD dose not increase the photosynthetic rate.

Uptake of atmospheric HTO into TFWT

The uptake of atmospheric HTO into TFWT was higher in rice plant than in Chinese cabbage while TFWT concentrations in both crops decreased rapidly after exposure. The reason for this may be that Chinese cabbage leaves are thicker than rice plant leaves and HTO are diluted by a larger volume of water. In comparison, the TFWT concentration in wheat leaves are about half the mean HTO concentration in the air humidity in case of an exposure under the light condition of about 600 μmol $m^{-2}s^{-1}$ (Table 2). In the rice plant experiment, HTO concentration in the irrigation water decreased very slowly while the HTO concentration in the top 1.4cm soil increased with time. One day later, tritium concentration in irrigation water and soil are very close (Fig. 5). In the Chinese cabbage experiment, the HTO concentration in the top 1.5cm soil decreased for one hour after exposure as rapidly as the TFWT concentration and thereafter a slower decrease occurred(Fig. 6). Tritiated water vapour from the atmosphere may directly enter the plants through the stomata. They occur in all green plant parts but the number of stomata per area is the highest in leaves. Tritiated water which has been deposited

Table 3. Specific activity ratio(SAR) in rice plants experiment.

Time after start of exposure(hr)	TFWT concentration (Bq/ml)	OBT concentration (Bq/ml combustion water)	SAR (OBT/TFWT)
1.0	134,000	770	0.0057
1.5	76,200	785	0.0103
2.0	32,800	970	0.0296
4.0	3,400	678	0.1994
6.0	1,200	550	0.4583
25	790	330	0.4177
97	630	157	0.2492
625(★)	73	136	1.8630
913(★)	227	299	1.3172

(★) : new stems

to soil may also be taken by the roots and the tritium concentrations in the plant are the contribution from the both sources [9,10].

Formation of OBT

Fig. 7 shows that the initial OBT concentration is relatively higher in Chinese cabbage (0.9%) than in rice plant (0.6%). The reason for this may be the higher light intensity during the Chinese cabbage experiment and the resulting higher rate of photosynthesis. The OBT formation in rice plant and Chinese cabbage are much lower compared to wheat leaves. The initial relative OBT concentration of wheat leaves was 1.4% under comparable light conditions [9]. Those lower values for rice plant and Chinese cabbage are mainly due to the fact that their samples contained stems which are less active in OBT formation. In both experiments, the OBT concentrations increased for the first 1 hour after HTO exposure because the TFWT concentration was still high enough for production of OBT. In the rice plant experiment, the OBT concentration decreased in the second 1 hour after HTO exposure and one day later it reached 43% of the initial OBT concentration. In the Chinese cabbage experiment, the OBT concentration increased further till 5 hours after HTO exposure and one day later it decreased to 48% of the initial OBT concentration. In order to compare the OBT formations under different light conditions and different tritium concentrations in the atmosphere, all OBT concentrations(Bq/ml) have been related to the TFWT concentrations in leaves of the first measured sample at the end of HTO vapour supply into the exposure box. OBT formation from HTO is only partly correlated with photosynthesis since there are tritium incorporation processes independent of light [11].

Specific activity ratio

The rate of tritium incorporation can be estimated by comparing the specific activities of the component and its precursor. The dose relevancy of studies is introduced by the difference in the concentration of tritium whether appearing in tissue or in organic molecules [12]. The specific activity ratio (R value) is defined as the value of organically bound tritium to tissue free water tritium. Most of R values in rice plant experiment were below 1.0 (Table 3). It means that tissue free water tritium concentration was higher than organically bound tritium after HTO exposure. But the ratio in the new plant which came out from contaminated pot was estimated to be above 1. This value was obtained about 1 month after HTO exposure. Chinese cabbage shows a similar trends to rice plant except higher R values(Table 4). For the purpose of tritium contamination indicators, it would seem practical to measure the ratio of HTO to OBT in the plants or animals. The ratio of HTO to OBT can provide not only an estimate of OBT dose, but also may serve as a possible marker of tritium source of contamination.

Tritium concentration of soil depths

After HTO exposure, tritium concentrations in soil water were measured for the whole depth of soil at intervals of about 3 cm. The soil of rice plant experiment was saturated with irrigation water until last sampling after HTO exposure while water content of soil in Chinese cabbage experiment was dependent on the weather condition such as rain, moisture etc. The profiles show two distinct tendencies (Fig. 8, 9). Tritium concentration increased with depth from the surface to the middle layer and then decreased up to the bottom of pot. This suggests that behaviour of HTO in the soil is similar to water behaviour and that it is also affected by diffusion, precipitation and time. The effective diffusion should be determined by the environmental conditions of the experiment. During the evaporation process, HTO diffuse into deeper soil layer because of the concentration gradient, while the top of layer loses activity to the atmosphere [13]. These results shows typical tritium diffusion in different kind of agricultural soil.

CONCLUSIONS

It is difficult to have a constant exposure condition in the field experiment because of differences in the velocity of HTO evaporation. Tritium concentration in the exposure box was dependent on external environmental conditions. The shape of plant leaves were related to the basic metabolism of plant and there was a

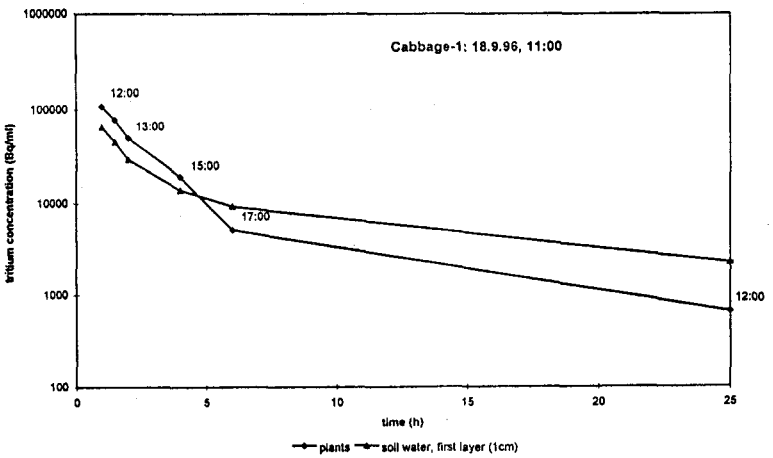

Fig. 6. Tissue free water tritium concentrations of Chinese cabbage and soil water at different times after HTO exposure.

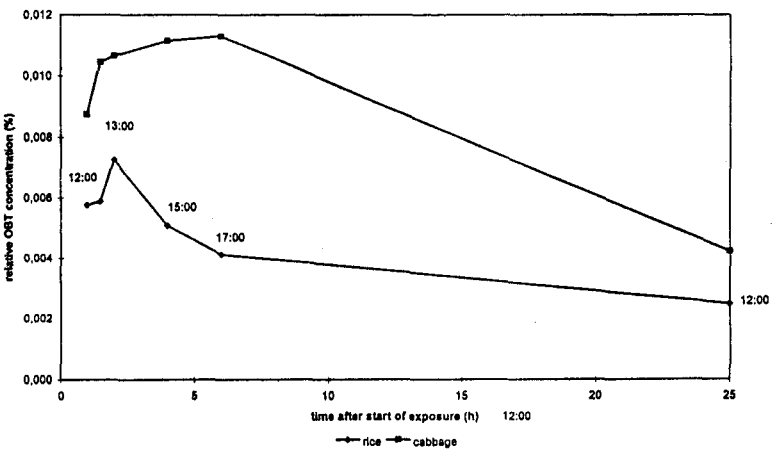

Fig. 7. Relative organically bound tritium concentration of rice plant and Chinese cabbage after HTO exposure.

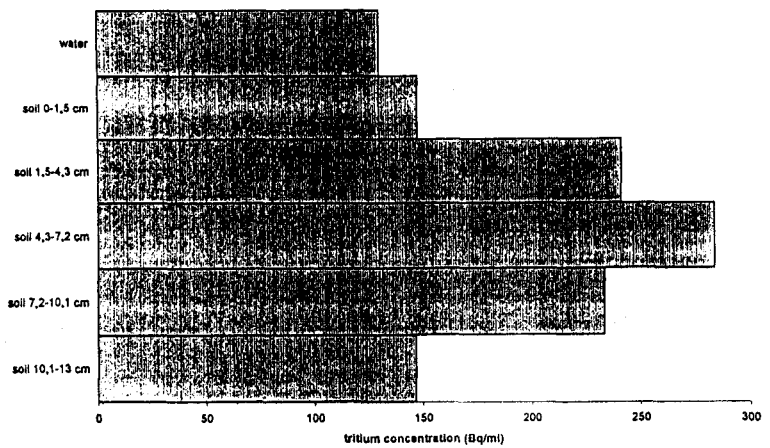

Fig. 8. Depth profile of tritium concentration in soil water of rice experiment after HTO exposure.

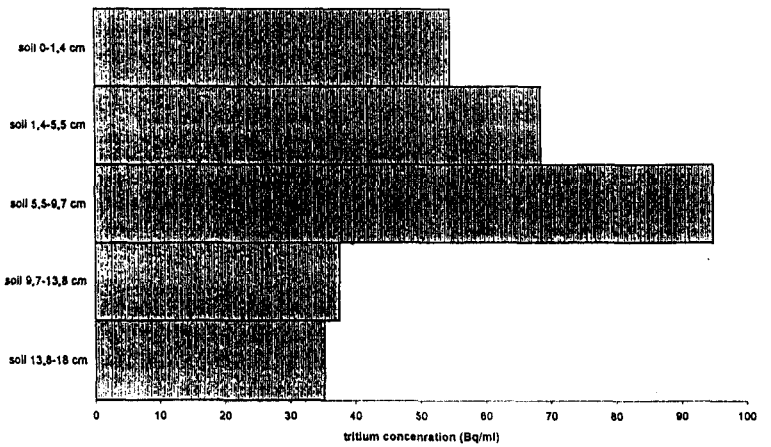

Fig. 9. Depth profile of tritium concentration in soil water of Chinese cabbage experiment after HTO exposure.

relationship between assimilation rate and light intensity. The uptake of atmospheric HTO into TFWT was higher in the rice plant than in Chinese cabbage and the decrease in TFWT concentration after HTO exposure was more rapid in the former than in the latter. The initial relative OBT concentration was higher in Chinese cabbage than in rice plant because the higher light intensity increased the rate of photosynthesis. The specific activity ratio can provide not only an estimate of OBT dose but also may serve as a possible marker of tritium contamination. Behaviour of HTO in the soil was similar to water behaviour and it seems to be affected by diffusion, precipitation and time.

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