https://doi.org/10.52676/1729-7885-2024-2-110-119
УДК 633/635:546.11.027*3:631.438.2:504.5 (574.41)

STUDY OF AERIAL TRITIUM UPTAKE BY VEGETABLE CROPS UNDER CHAMBER AND FIELD CONDITIONS

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Under laboratory (in a chamber) and full-scale conditions (at the former Semipalatinsk Test Site), pepper and eggplant were exposed several times to tritium in the form of HTO at different growth stages. In chamber experiments, tritium activity concentration in the free water (TFWT) of plants' leaves during exposure increased, and under full-scale conditions, it was marked by an unsteady dynamic. TFWT activity concentration in leaves of both crops was 1–2 orders of magnitude higher than in stems and fruits. Values of $TFWT/HTO_{air}$. showed that TFWT activity concentration at the end of exposure reached equilibrium only in leaves. Both in full-scale and chamber experiments, tissue free water was noted to be enriched with tritium compared to the ambient air ($TFWT/HTO_{air}$ >1). In the post-exposure period, FWT activity concentration in both crops quickly dropped on the first day (over 90–95%). In the next 2 weeks (336 h), a reduction in TFWT was markedly slowing down. In all experiments, activity concentration values of organically bound tritium (OBT) in pepper and eggplant leaves are 1–2 orders of magnitude lower than TFWT. OBT activity concentration in both crops on the first day after exposure was marked by both a positive and negative dynamics. After 2 weeks (336 h) following the exposure, the loss of OBT was 60–95%. A close correlation relationship was established between TFWT activity concentration in leaves and HTO in the air ($r = 0.73$; $p<0.05$), and a moderate one – between TFWT activity concentration in leaves and the air humidity ($r = 0.54$; $p<0.05$). No significant correlation relationship was revealed between OBT activity concentration in leaves and environmental factors (photosynthetically active radiation, temperature, relative humidity). Findings showed that a possible contribution by organically bound tritium to the annual average internal exposure dose on consumption of crop products exposed to a short-term aerial contamination by HTO will be negligible. Data from full-scale experiments, taking into account the impact by actual climatic factors, could be used to test regional models of tritium transport by air in the "air-to-agricultural plant" system.

Keywords: *tritium, tritium oxide, tissue-free-water tritium, organically-bound tritium, crops, aerial uptake, chamber and field experiment.*

INTRODUCTION

Tritium is the radioactive isotope of hydrogen. It disintegrates to ^3He with emissions of a low energy (≤18.6 keV) β-electron and an antineutrino at a half-life of 12.323 years [1] Currently, the main source of tritium in the environment is routine release from nuclear fuel cycle facilities (NFC), in particular nuclear power plants [2–4]. It is expected in the future nuclear fusion power plant will use tritium as fuels [1]. As a consequence, one of unavoidable problems for the realization of a nuclear fusion reactor in future will quantitatively evaluate and prediction of biological incorporation and transfer of tritium in plant as nature ecosystem as agriculture ecosystems. The base forms of tritium releases are tritiated hydrogen (HT, T_2) and tritiated water or tritium oxide (HTO, T_2O) [2, 4, 5]. The tritiated hydrogen is not assimilated by aerobes and it is oxidized to tritium oxide (HTO) in the environment [2]. Thus, the tritium oxide or tritiated water is the most common inorganic form of isotope existence, which has exceptional migration activity and bioavailability as a result of the identity of chemical properties with the ordinary water molecule [3–8]. The tritiated water is easily enters into the trophic structure of the ecosystem, the final link of which can be a person. Plants play a key role in these processes due to their ability is incorporated tritium into primary organic matter directly during photosynthesis [5, 8–10].

The interest of organically bound tritium (this definition will be used further on in the article as OBT) formation and transfer, especially by crops, significantly increased because it can translocate into edible parts [8, 9, 10–14]. Besides, the OBT has more resistance in human body compared to tritiated water [15–18]. Also, it should be taken into consideration, that dose coefficient of OBT is almost by a factor of 3 higher than of HTO [15–17]. There is necessary for more reliable risk assessment of accidental tritium releases is emphasized for nuclear energy future [19]. The development of a standard conceptual model for accidental tritium releases is difficult, because tritium transfer to crops can depends on changing meteorological condition, plant physiological processes and local consumption [3, 5–9, 13]. The robust assessment of possible radiological risks of crop contamination with tritium as a result of its accidental release must be based on the appropriate experimental database. The aerial pathway of tritium to a plant is complex compared to the root uptake, and this requires a detailed study to understand this phenomenon [3, 7, 8]. The literature contains a lot of data obtained either under field or under laboratory conditions [22–26] on the aerial uptake of tritium by cereal crops at different grain-ripening stages. Diabaté and Strack [23] studied the uptake of tritium by wheat after a short-term exposure to HTO vapor under

laboratory conditions. Atarashi-Andoh et al. [24] conduct release experiments using deuterium as a substitute for tritium and exposed potted rice plants to deuterated water vapor. Choi et al. [25] conducted experiments with rice outside at several different growth stages from booting to harvesting and estimated that most of the human radiation dose due to ingestion of rice is caused by OBT. There also is data on the tritium uptake in grass vegetation [27] and leafy vegetables [28, 29, 30]. Research presented was conducted to comparatively evaluate the aerial tritium uptake by crops (pepper and eggplant) at different growth stages under laboratory and field conditions simulating an accidental release and the rate of tritium loss after a long-term exposure.

OBJECTS AND METHODS RESEARCH

Plants cultivation. Pepper (*Capsicum annuum L.*) and eggplant (*Solanum melongena L.*) were used in experiments. These crops are typical vegetables which are cultivated all over the Republic of Kazakhstan. For exposure experiments under laboratory and field conditions, plants were cultivated in plastic pots (V=35 L) with light chestnut loamy soil. Dry seeds similar in size and weight were sown at the rate of 2–3 seedlings per 1 pot. The plants were grown in a greenhouse until the beginning the different growth stages (exponential growth, flowering and maturity). Crops were watered with distilled water maintaining an optimal humidity of 60% of full soil moisture capacity. Appropriate amounts of the composite fertilizer were supplied as required during the plant growth. Also, disease and insect controls were carried out as required.

Experimental technique. Plant exposure started between 9:30 a.m. and 10:00 a.m. and ended between 16.00 p.m. and 18.00 p.m. Thus, during the exposure in all experiments, the diurnal rhythms of plants matched. This condition is important, since most physiological processes in plants, in particular, resistance of stomata and photosynthetic activity, are linked to daily biorhythms [32].

Plant exposure lasted from 6 to 8 hours. During the exposure, the soil in the pots was covered with polyethylene film to prevent HTO from being diffused into the soil and, accordingly, from being uptaken by roots. After exposure, the plants were left to grow naturally in a well-ventilated room and kept for 14 days while being irrigated with distilled water.

Plants (leaves, stems, fruits) during the exposure were sampled at 2 h intervals, 1 h, 24 h and 14 days after the exposure. All plant samples were collected in 3–4 replications. Once collected, plant samples were divided into leaves, stems and fruits. The weight of each plant sample averaged 100–150 g. To prevent the loss of tritium in the field, plant samples were immediately packed in zip bags and stored in a portable freezer at −20 °C. Air samples were collected at 1–2 h intervals using a tritium collector "OS 1700" (AMETEK, USA).

An experiment under laboratory conditions was conducted in a metal-plastic chamber. The chamber size is 2500×1500×1200 mm. The chamber is equipped with a phyto-lighting system. From the perspective of energy efficiency, the illumination level in the chamber corresponded to plant needs in the photosynthetically active radiation (PAR) [33]. About 20–25 plants of each crop were exposed to HTO vapor in the chamber. To do so, a tray containing water with a tritium concentration of $(5.6 \cdot 10^4)$ Bq·l^{-1} was placed at the bottom of the chamber. To continuously measure the concentration of HTO vapors in the air, a tritium collector sampling hose was placed in the chamber through a special technical hole in the side wall of the chamber.

For a better reproduction of the real impact of environmental conditions and comparative analysis with laboratory results, similar experiments were additionally carried out in the territory of the Semipalatinsk Test Site (STS) at the former technical sites "Degelen". This technical site characterized by high levels of tritium contamination in the surface air near the radioactive tunnel watercourses due to underground nuclear tests. The maxima of the tritium concentration in the air recorded in the chosen area of experiments conducted [34, 35] were 1400 and 700 Bq m^{-3}. The 30 pots with pre-cultivated crops at different stages of growth were placed near radioactive tunnel watercourses.

During the experiments, temperature, relative humidity, and atmospheric pressure were recorded at 1 h interval using a thermal hygrometer (IVA-6, Russia). The light intensity was measured using a spectrometer (TEK, Russia).

Extraction of tissue-free water tritium in plant samples. Free water of tissue was extracted from plant samples using a special device [36]. Samples of plant were put into transparent box, which had a cover in the form of a cooling vessel. In this device cold water was used as a cooling agent. By natural evaporation of the plant sample, free water condensate was obtained from the plant tissues. The obtained condensate corresponded to tritium as a tritiated water named "tissue-free water tritium" (this definition will be used further on in the article as TFWT). The extracted TFWT was transferred to a counting polyethylene vial. The volume of condensate varied from 10 to 15 ml. The TFWT activity concentration was measured by liquid scintillation spectrometry.

Combustion method for determining organically-bound tritium in plants samples. After free water was extracted, the plant samples were dried in a laboratory drying oven (BINDER ED 53, Germany) at a temperature of 70–80 °C and powdered. About 2.0 g of the dry sample combusted with a "Sample Oxidizer" (PerkinElmer Model No. 307, USA). Water obtained after combustion was collected in a tritium counting vial. The OBT activity concentration was measured by liquid scintillation spectrometry.

Liquid scintillation counting. Tritium activity concentration was measured by liquid scintillation spectrometry using a "QUANTULUS 1220" spectrometer (Perkin Elmer, USA) [37]. Prior to the measurement, samples

were filtered to remove mechanical impurities, then a 3 ml aliquot was collected into a 20 ml plastic vial and an Ultima Gold LLT scintillation cocktail for natural samples (the registration efficiency for tritium in the 0-18 keV range of about 60%) was added at a ratio of 1:4 ("sample-scintillator" ratio). The measurement time was at least 120 minutes, the beta spectra were processed and the activity concentration of tritium was calculated using the program "Quanta Smart". The minimum detectable activity of tritium was from 4 to 7 Bq L^{-1} depend on tritium form.

The volumetric activity of tritium in each air sample was determined as the arithmetic mean from measurements of 3 counting samples prepared from the initial sample.

RESULTS AND DISCUSSION

Exposure conditions. The relative humidity, temperature, and light intensity were recorded during exposures every hour. The light intensity given as measured in PPFD (density of the photosynthetic photon flux). Table 1 provides data on the meteorological conditions during each exposure.

The mean values of air temperature ranged from 30 to 33.4 °C in chamber experiments. In the field experiment, the mean temperature was 27.3 °C. Relative air humidity in most cases varied from 61 to 90%.

Aerial tritium uptake by pepper and eggplant

TFWT in pepper and eggplant. Table 2 provided the HTO activity concentration in air moisture and in the free water of vegetables during each exposure under chamber and field conditions.

According to the Tables 2 and 3, the HTO concentration increased very rapidly towards the end of exposure both under chamber and field conditions. The maximum values of the tritium activity in air were observed in the chamber experiments. Throughout the chamber exposure, HTO concentration varied from (1303±130) to (7366±1100). During the field experiment, HTO concentrations varied from (341±57) to (1107±108) Bq·l^{-1}. In general, the concentration of HTO in the air of the chamber was 1.5–21 times higher than in the field.

As shown by Table 3, the activity concentration of tritium in the free water of vegetables increased at the end of exposure compared to the initial activity concentration (2 h) in chamber experiments.

Table 1. Meteorological conditions during exposures

Cod of exposure	Temperature, °C		Relative humidity, %		PPDF, μmol s^{-1} m^{-2}	
	Range	Mean±SD (n)	Range	Mean±SD (n)	Range	Mean±SD (n)
Camber experiment						
E_{EG}	24.1–35	30±5 (6)	65–88	78.7±6 (6)	851–902	907.3±21 (6)
E_{F+M}	22.8–23	22.7±4,5 (6)	89–92	91±5.5 (6)	900–903	901±24 (6)
P_F	23.4–41.7	33.4±5 (6)	61–90	76±6 (6)	66–143	101.3±32 (6)
P_M	23.4–41.7	33.4±4 (6)	82–91	90.1±8 (6)	27–435	192±65 (6)
Field experiment						
E_{VG}, E_{F+M}, P_{F+M}	23–29	27.3±3,5 (8)	65–80	68.3±6 (8)	6607–2554	3680±786 (8)

n – number of cases;
Note to Tables 1–9: E – eggplant; P – pepper; EG – exponential growth; F – flowering; M – maturation.

Table 2. Mean values of tritium activity concentrations in experimental samples in the chamber experiments

Cod	Part	Tritium activity concentration, Bq·l^{-1}								
		Exposure period, h						Post-exposure period, h		
		2		4		6		1	24	336
		TFWT	HTO	TFWT	HTO	TFWT	HTO	TFWT		
E_{EG}	leaf	1600±200	7366±1100	2500±400	3492±525	2800±400	3810±571	2000±300	590±90	156±23
	stem	270±40		590±90		920±140		920±140	430±65	214±32
E_F	leaf	2700±400	1576±234	3300±500	3698±560	4100±600	4832±730	2600±400	220±35	230±35
	stem	160±25		270±40		220±22		190±29	33±5	65±10
P_F	leaf	3000±400	3141±470	4600±700	3672±551	5200±800	4271±639	3700±600	690±100	—
	stem	210±30		240±40		420±60		200±35	121±18	—
E_M	leaf	1100±200	1576±234	2400±400	3698±560	4500±650	4832±730	2300±300	350±60	260±40
	stem	84±12		250±47		390±60		340±50	46±7	56±8
	fruit	38±6		107±16		330±50		32±5	240±34	120±16
P_M	leaf	2900±400	1386±153	5000±750	1329±124	8300±1150	1303±131	4200±650	840±125	—
	stem	176±26		470±70		960±140		410±60	107±16	—
	fruit	151±24		260±40		650±100		640±100	470±70	—

Note to Tables 2–8: "—" – date is not obtained

Table 3. Mean values of tritium concentrations in experimental samples in the field experiment

Cod	Part	Tritium activity, Bq·l⁻¹										
		Exposure period, h								Post-exposure period, h		
		2		4		6		8		1	24	336
		TFWT	HTO	TFWT	HTO	TFWT	HTO	TFWT	HTO			
E_{EG}	leaf	1200±180	341±57	310±40	605±61	315±50	1107±108	—	994±99	295±40	95±14	54±7
	stem	115±17		89±15		95±14		—		85±13	71±12	59±8
E_F	leaf	365±50		340±50		555±100		1100±200		—	—	—
	stem	24±4		18±3		16±3		43±6		—	—	—
P_F	leaf	—		635±90		610±90		480±750		425±60	110±15	132±22
	stem	—		28±4		33±5		34±5		30±4	51±8	96±22
E_M	leaf	—		1400±200		1200±170		1200±200		530±80	87±13	10±2
	stem	—		57±8		33±4		74±11		56±8	52±7	8±1,2
	fruit	—		48±7		70±10		110±16		80±13	70±10	45±5
P_M	leaf	166±25		220±34		320±45		250±35		360±55	86±12	4±0,8
	stem	17±3		40±6		18±3		22±4		35±5	42±6	4±0,8
	fruit	17±3		40±5		18±3		22±4		35±5	42±6	4±0,8

Table 4. Concentration ratio TFWT/HTO_{atm} at the end of exposure

Cod	C_{HTOatm} at the end of exposure, Bq·l⁻¹ (mean±SD)	Concentration ratio C_{TFWT} (Bq·l⁻¹)/C_{HTOatm} (Bq·l⁻¹) (mean±SD)		
Chamber experiment				
		leaves	stems	fruits
E_{EG}	3810±500	0,7±0,1	0,20±0,02	no
E_F	4832±620	0,8±0,2	0,10±0,003	no
P_F	4271±640	1,2±0,2	0,10±0,01	no
E_M	4500±650	0,9±0,1	0,08±0,002	0,07±0,003
P_M	1303±620	6,4±0,2	0,70±0,1	0,50±0,1
Field experiment				
E_{EG}	315±50	0,3±0.05	0,05±0,01	no
E_F	1100±200	0,5±0,1	0,03±0,003	no
P_F	480±750	0,5±0,1	0,03±0,005	no
E_M	1200±200	1,2±0,2	0,07±0,01	0,11±0,02
P_M	250±35	0,3±0,04	0,02±0,004	0,02±0,004

"no" – no fruits at this growth stage.

According to data in Table 3, during field exposure the dynamic of the TFWT was not as uniform as in chamber experiments.

During the P_F, E_M, P_M field exposure, the TFWT activity concentration even decreased by a factor of 1–3 compared to the initial concentration (2 h) exposure. This may be due to environmental factors such as air movements, vapor pressure deficit, atmospheric CO_2 levels and relative humidity influencing the conductivity of stomata under field conditions and consequently the exchange of HTO between the plant and ambient air.

The TFWT activity concentration in pepper and eggplant leaves were 1–2 orders of magnitude higher than in the stems and fruits during exposures. Only at the exponential growth stage the difference was by a factor of 5–8. This fact is certainly attributed to the isotope entry through the stomata. The obtained data is in agreement with the results of rice exposed to atmospheric HTO vapor under semi-outdoor conditions [25], and wheat exposed in the box [23]. In the field experiments conducted by Kline and Stewart [27] the TFWT activity concentration in grass leaves was also an order of magnitude higher than in other plant parts during the exposure to atmospheric HTO.

Table 4 shows the observed ratios between the tritium concentrations of tissue water in different plant parts and of the air humidity (HTO_{atm}) at the end of the exposure under chamber and field conditions.

According to obtained for values of TFWT/HTO_{atm}, tritium concentration in free water was reached to steady-state at the end of exposure only in leaves. The TFWT/HTO_{atm} ratio varied from 0,3 to 6,4. In the stems and fruits the concentration of the isotope did not reach to steady-state at the end of 6-8-hour exposure. This may be due to the surface relative to mass is much smaller in stems and fruits than in leaves leading to lower TFWT concentrations [23].

In chamber (P_M, P_F) and field (E_M) experiments the values of TFWT/HTO_{atm} were a higher than 1.0 because the tritium concentration in the free water of leaves was higher than in the ambient air. The enrichment of the free water of leaves with tritium can occur in transpiration when the liquid phase passes into steam. When crossing the liquid-gas phase interface, "light" water molecules are preferably released due to the difference in vapor pressure between H_2O and HTO when the ambient temperature rises [38]. As a result, there is a "weighting" of free water of tissues by concentrating HTO. During box experiments with wheat the TFWT concentration in leaves is dominated by the tritium concentration in the atmosphere only under strong light conditions and with relative humidity around 90% [23]. The literature also contains data on the enrichment of free water by other heavy isotopes concentrations of which exceeded the level in the ambient air [39, 40]. The phenomenon of enrichment in the free water of leaves with tritium can lead to a more active involvement of the isotope in metabolic processes and, as a consequence, this can lead to fractionation of isotope in plant organic matter. This phenomenon must be taken into account especially in the case of plants with a rapid rate of photosynthesis.

According to Table 4, most of HTO mean concentrations under chamber conditions were 1 order of magnitude higher than the ones under field conditions. However, the values of TFWT/HTO_{atm} for E_{EG}, E_M, P_F and P_M in chamber and field experiments indicate that the concentration of the radionuclide in the free water of plants might not only depend on the level of tritium contamination. In these variants of exposure seems the vary of the TFWT concentration in the pepper and eggplant to be primarily due to the difference between in the stomatal conductivity of leave crops under influence of environmental factors [41].

As shown by Tables 2 and 3, after the end of every exposure, the TFWT concentration in plant parts decreased at a rapid rate for the first day, at a much slower rate for the following 336 h. Overall, 336 h after exposure, the loss of tritium in the free water of leaves was on average more than 90–95% compared to the level at the end of exposure. The reason for this is the dilution of the intercellular sap with pure water absorbed by the plant through the root, the loss during transpiration and the inclusion of TFWT in metabolic processes. the presented data are in good agreement with the results of rice experiments [25]. After the end of every exposure, the TFWT concentration in each rice part decreased at a rapid rate in the first several hours, at a much slower rate for the following 300–500 h.

OBT in crops. The Tables 5 and 6 provide mean values of OBT activity concentration during experiments.

The OBT activity concentration in leaves increased only by a factor of 1–4 at the end of exposure compared to the initial concentration (2 h) exposure. This is due to the fact the organically bound form of the radionuclide is a result of biochemical processes, which requirement more time than diffusion of HTO through stomata [10, 11, 13]. Furthermore, OBT formation depends on the plant physiological state and rate of photosynthesis [10].

All values obtained for OBT in pepper and eggplant leaves were 1–2 orders of magnitude lower than TFWT in both chamber and field experiments. This reflects the fact that about 90% of the organically bound form of the isotope is formed in leaves during photosynthesis [6, 9, 16]. The results of experiments with wheat [23], rice [25], Chinese cabbage [42] exposure to HTO also showed 1 showed OBT being 1-3 orders of magnitude higher in leaves.

Table 5. Mean values of OBT activity concentrations in crops in chamber experiments

Cod of exposure	Part	OBT activity concentration, Bq·l^{-1}					
		Exposure period			Post-exposure period		
		2 h	4 h	6 h	1 h	24 h	336 h
E_{EG}	leaf	66±11	96±14	161±23	234±30	176±24	79±12
	stem	39±9	69±13	146±22	232±29	98±17	58±11
E_F	leaf	92±15	125±22	98±17	145±25	92±17	50±10
	stem	18±5	28±7	35±8	47±12	57±12	15±4
P_F	leaf	80±12	117±18	170±25	162±25	100±17	—
	stem	17±7	50±8	42±7	33±5	55±8	—
E_M	leaf	33±8	75±12	75±13	70±13	62±10	22±5
	stem	38±7	33±10	17±5	40±10	18±5	12±4
	fruit	17±5	25±7	28±8	27±7	50±12	28±5
P_M	leaf	35±8	38±7	78±12	72±12	50±10	—
	stem	10±3	4±2	13±4	48±10	40±8	—
	fruit	50±9	27±7	30±7	60±12	50±10	—

According to Tables 6 and 7, the dynamics of OBT concentration during the first hour and 24 hours after exposure did not show a clear pattern. In most cases, however, there was a decrease in OBT concentration. After 336 hours following the end of exposure, the OBT concentration decreased more slowly compared to the TFWT concentration in the parts of pepper and eggplant. This indicates that after exposure the conversion of leaf TFWT into organic matter and outflux of tritium assimilates continues. Observed small peaks of OBT concentration in pepper at the maturation stage (E_M) 24 hours after the end of exposure, are also attributable to these reasons. On the other hand, a decrease in OBT because of newly formed organic matter occurs after exposure in which tritium was not included. In the experiments conducted by Choi et al. [25] similar dynamic of OBT concentrations in different rice exposed to atmospheric HTO was found. It should be noted that after exposure the loss of organically bound tritium occurred slowly for both crops, once translocated to fruits. A similar pattern was established in experiments with wheat by Diabate´and Strack [23], and with rice by Choi et al. [25].

Since the concentrations of HTO in air varied from experiment to experiment (Table 2, 3), findings were normalized to make it easier to compare trends between calculations and observations in different experiments. All OBT concentrations (Bq l^{-1}) were related to TFWT in leaves concentrations measured at the end of exposure. It should be noted, in the presented study the OBT-to-TFWT ratio is not the so-called "R-value" or specific activity ratio (SAR) [5, 8, 9, 13, 22] cited in the literature. The values of OBT-to-TFWT ratio for all parts of pepper and eggplant are summarized in Table 7.

The low values of OBT-to-TFWT obtained in this study are apparently attributed to a short-term exposure. The OBT concentration ratio in this study are in good agreement with the results of the short-term exposure to HTO vapor presented in the literature [25, 35, 40].

Table 6. Mean values of OBT activity concentrations in crops in the field experiment

Cod of exposure	Part	OBT activity concentration, Bq·l^{-1}						
		Exposure period				Post-exposure period		
		2 h	4 h	6 h	8 h	1 h	24 h	336 h
E_F	leaf	22±6	25±7	12±5	50±10	—	—	—
	stem	6±3	12±5	15±5	20±5	—	—	—
P_F	leaf	—	—	—	—	—	—	33±7
	stem	—	—	—	—	—	—	35±7
R_M	leaf	—	—	—	—	—	—	—
	stem	—	—	—	—	53±10	—	—
	fruit	—	—	—	—	25±7	—	—
P_M	leaf	12±4	37±10	30±7	33±8	43±8	67±15	10±4
	stem	8±1	13±5	7±4	18±7	15±5	12±5	15±5
	fruit	—	9±4	23±7	33±8	17±5	32±8	33±8

Table 7. Concentration ratio OBT/TFWT$_{leaf}$ at the end of exposure

Cod of exposure	C_{TFWT} in the leaves at the end of exposure, Bq l^{-1} (mean±SD)	Concentration ratio C_{OBT} (Bq l^{-1})/$C_{TFWT\ leaves}$ (Bq l^{-1}) (mean±SD)		
Chamber experiment				
		leaves	stems	fruits
E_{EG}	2800±400	0.06±0.002	0.30±0.01	no
E_F	4100±600	0.01±0.001	0.20±0.01	no
P_F	5200±800	0.03±0.001	0.01±0.003	no
E_M	4500±650	0.02±0.001	0.01±0.003	0.01±0.001
P_M	8300±1150	0.01±0.003	0.01±0.001	0.05±0.003
Field experiment				
E_{EG}	315±50	—	—	no
E_F	1100±200	0.05±0.01	0.02±0.003	no
P_F	480±75	—	—	no
E_M	1200±200	—	—	—
P_M	250±35	0.10±0.02	0.10±0.03	0.10±0.03

"no" – no fruits at this growth stage

Table 8. Correlations matrix

Variable	No.	Spearman rank order correlations (r_s)					
		1	2	3	5	6	7
$C_{TFWT\ (leaf)}$ (Bq l^{-1})	1	1.00	−0.10	0.73*	0.21	0.12	0.54*
$C_{OBT\ (leaf)}$ (Bq l^{-1})	2		1.00	0.03	−0.22	0.12	−0.23
C_{HTOair} (Bq l^{-1})	3			1.00	0.45*	0/34*	0.33*
t (°C)	5				1.00	0.11	−0.35
PPDF (μmol s^{-1} m^{-2})	6					1.00	−0.23
φ (%)	7						1.00

* Correlations values highlighted are statistically significant at p<0,05

According Table 4, the concentrations in the leaves of pepper and eggplant during chamber experiment were significantly higher than ones under field conditions. Despite this the estimated OBT-to-TFWT ratios did not show any clear relationship between OBT formation and the level of TFWT in leaves. One of the possible reasons may a limitation of photosynthesis under chamber conditions by a likely impoverishment of CO_2 because of it plays one of key role as limit factor [10, 42]. The results of wheat box experiments [23] also showed the limitation of photosynthesis. On the other hand, the rate of photosynthesis depends on light intensity [9, 10, 13]. Under field conditions the values of PPDF were one order of magnitude higher than in the chamber (Table 1).

As shown by Table 4, the concentration ratio $OBT/TFWT_{leaves}$ did not show clear differences related to the growth stage neither for pepper nor eggplant. In paper by Atarashi-Andoh et al. [23] found no differences in the concentration of the isotope in rice at the grain growth stages during exposure using deuterium as an analogue of tritium. But it should be noted, in the rice experiments conducted by Choi et al. [25] ear OBT concentration and the seed and chaff OBT concentrations at harvest were greatly affected by grain-ripening stage on which exposure occurred.

Dependence between the tritium activity and environmental factors. The assessment of the relationship between the variables (HTO_{air}, $TFWT_{leaf}$, and OBT_{leaf}) and the environmental factors is provided in the correlation matrix (Table 8). For the analysis, only the values of the concentrations of TFWT and OBT in the leaves were used, since more than 90% of tritium is incorporated into organic matter during photosynthesis in leaf chloroplasts [5, 8–10].

According to Table 8, there is a positive strong correlation relationship ($r_s = 0.73$) between $TFWT_{leaf}$ and HTO_{air} activity concentration. At the same time, between $TFWT_{leaf}$ activity concentration and air humidity the positive moderate correlation relationship ($r_s = 0.54$) was established. In turn, the HTO_{air} activity concentration in air weak correlates with air temperature ($r_s = -0.45$), PAR intensity ($r_s = 0.34$), and relative humidity ($r_s = 0.33$).

No statistically significant correlation was established between the OBT activity concentration in leaves and the tritium activity concentrations in free water, as well as the air HTO. This result is also confirmed by the values of the OBT-to-TFWT ratio for leaves obtained in chamber and field experiments (Table 7). Also, the results of the correlation analysis did not reveal a dependence of the concentration of OBT in leaves on environmental factors, such as PAR, temperature, and relative humidity (Table 8). This is explained by the key role of physiological aspects in the intensity of photosynthesis. The ecological aspect of the influence of temperature and lighting conditions on photosynthesis is related to daily and seasonal cycles only. The most characteristic feature is the single-peak shape of the daily photosynthetic gas exchange curve, with a pronounced midday maximum on a clear day and a gently domed maximum on a cloudy day [44].

CONCLUSION

A series of experiments with pepper and eggplant plants at different growth stages were conducted under laboratory (in the chamber) and field conditions (in the former STS territory). Concurrent experiments with the same crops gave a unique opportunity to compare data obtained under simulated and natural conditions.

The $TFWT_{leaves}$ activity concentration increased at the end of exposure in chamber experiments. During the field exposure, the concentration of $TFWT_{leaves}$ showed unstable dynamics. The level of $TFWT_{leaves}$ activity in both crops was 1–2 orders of magnitude higher than in other plant parts. The TFWT-to-HTO_{atm} ratios indicated that the TFWT activity concentration reached a steady-state only in leaves at the end of exposure. In some variants of chamber and field experiments TFWT activity concentration dominated to the level of isotope in the ambient air. The phenomenon requires more attention to risk assessment, especially, in cases where plants have a higher rate of photosynthesis. In other plant parts the TFWT activity concentration did not reach a steady-state during exposure. After exposure the TFWT loss in leaves averaged more than 90–95% compared to the end of exposure.

After short-term exposure the organically-bound tritium (OBT) concentration in both crops was only a small fraction of the one in tissue water. The OBT activity concentrations in pepper and eggplant leaves were 1–2 orders of magnitude lower than TFWT in all variants of experiments. The OBT/TFWT ratios did not show any clear relationship between OBT formation and the level of TFWT in leaves. The clear relationship between OBT

formation and TFWT concentration in leaves was not established. During post-exposure period the OBT activity concentration in both crops slowly decreased compared to TFWT. The OBT loss in all parts averaged in range 60–95% compared to the end of exposure.

A positive strong (r = 0.73) correlation was found between TFWT leaf and HTO air activity concentrations, and a positive moderate (r = 0.54) correlation was also found between TFWT leaf and air humidity. No correlation was found between the OBT concentration in leaves and environmental factors such as PAR (photosynthetically active radiation), temperature, and relative humidity.

The risks from OBT in crop products exposed to a short-term aerial contamination with the radionuclide at any development stage would be negligible. In case of testing regional models of aerial transfer of tritium in plants and incorporation of the radionuclide into organic matter, it would be best to use field experimental data that takes into account the influence of actual climatic factors.

Funding

This research was funded by the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan (BR21882185).

Acknowledgements

Authors appreciate the assistance from the staff of the branch 'Institute of Radiation Safety and Ecology' RSE NNC RK (Kurchatov, Kazakhstan) in radioecological research. Specific thanks to K.Y. Tomilov for translating the article, L.F. Subbotina for sample preparation.

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КАМЕРАДА ЖӘНЕ ТАБИҒИ ЖАҒДАЙДА КӨКӨНІС ДАҚЫЛДАРЫНЫҢ ТРИТИЙДІ АЭРОЗОЛЬМЕН СІҢІРУІН ЗЕРТТЕУ

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Зертханалық (камерада) және далалық жағдайларда (бұрынғы Семей сынақ полигонының аумағында) вегетативтік дамудың әртүрлі кезеңдерінде бұрыш және баклажан дақылдарымен бірқатар эксперименттер жүргізілді. Камералық эксперименттерде өсімдік жапырақтарындағы бос судағы тритийдің (БСТ) шоғырлануы экспозицияның соңына қарай айтарлықтай артты, ал далада тіпті БСТ 1–3 есеге төмендеді. Екі дақылдың жапырақтарындағы БСТ шоғырлануы сабақтар мен жемістерге қарағанда 1–2 есеге жоғары болды. БСТ/НТО$_{эсер}$ қатынасынан, БСТ шоғырлануы экспозицияның соңына қарай жапырақтарда ғана тұрақты күйге жеткенін көрсетті. Кейбір жағдайларда жапырақтардың бос суындағы тритийді қоршаған ауамен салыстырғанда байытылуы байқалады. Экспозиция кезінде сабақтар мен жемістерде БСТ шоғырлануы тепе-теңдікке жеткен жоқ. Экспозицияның соңына қарай жапырақтардағы ОБТ шоғырлануы 1–4 есе өсті. Эксперименттің барлық нұсқаларында ОБТ шоғырлануы БСТ-ге қатысты 1–2 есеге төмен мәндерге ие болды. ОБТ түзілуінің жапырақтардағы отв шоғырлануына, сондай-ақ вегетативті даму сатысына тәуелділігі анықталмаған. Экспозициядан кейінгі кезеңде өсімдіктерде БСТ шығыны 90-95%, ОБТ – 60-тан 95% дейінгі мәнді құрады.
Зерттеу нәтижелері тритийдің апаттық шығарындылары жағдайында қоғамдық қатерді жан-жақты бағалау үшін табиғи жағдайда алынған деректерді пайдаланған дұрыс екенін көрсетті. Сонымен қатар, зерттеу нәтижелерін аймақтық жағдайларда қолданыстағы тритий тасымалдау үлгілерін тексеру үшін пайдалануға болады.

Түйін сөздер: тритий, тритий оксиді, бос судағы тритий, органикалық байланысқан тритий, дақылдар, аэральды сіңіру, камералық және табиғи тәжірибелер.

ИССЛЕДОВАНИЕ АЭРАЛЬНОГО ПОГЛОЩЕНИЯ ТРИТИЯ ОВОЩНЫМИ КУЛЬТУРАМИ В КАМЕРЕ И НАТУРНЫХ УСЛОВИЯХ

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В лабораторных (в камере) и натурных условиях (на территории бывшего Семипалатинского полигона) была проведена серия экспозиций культур перца и баклажана тритием в форме НТО на разных стадиях роста. В камерных экспериментах удельная активность трития в свободной воде (ТСВ) листьев растений в ходе экспозиции увеличивалась, а в натурных условиях характеризовалась нестабильной динамикой. Удельная активность ТСВ в листьях обеих культур была на 1–2 порядка выше, чем в стеблях и плодах. Значения ТСВ/НТО$_{возд.}$ показали, что удельная активность ТСВ в конце экспозиции достигает равновесного состояния только в листьях. Как в натурных, так и в камерных экспериментах наблюдалось обогащение свободной воды тканей тритием по сравнению с окружающим воздухом (ТСВ/НТО$_{возд}$ >1). В пост-экспозиционный период удельная активность ТСВ в обеих культурах быстро снижалась в первые сутки (более 90–95%). В последующие 2 недели (336 ч) снижение ТСВ значительно замедлялось. Во всех экспериментах значения удельной активности органически-связанного трития (ОСТ) в листьях перца и баклажана на 1–2 порядка ниже по сравнению с ТСВ. Удельная активность ОСТ в обеих культурах в первые сутки после экспозиции характеризовалась как положительной, так и отрицательной динамикой. Через 2 недели (336 ч) после экспозиции потеря ОСТ составила 60–95%. Тесная корреляционная зависимость установлена между удельной активностью ТСВ в листьях и НТО в воздухе ($r = 0.73$; $p<0,05$), и умеренная – между удельной активностью ТСВ в листьях и влажностью воздуха ($r = 0.54$; $p<0,05$). Между удельной активностью ОСТ в листьях и факторами окружающей среды (фотосинтетически активная радиация, температура, относительная влажность) достоверная корреляционная зависимость не выявлена.
Полученные результаты показали, что возможный вклад органически-связанного трития в среднегодовую дозу внутреннего облучения при употреблении растениеводческой продукции, подвергшейся кратковременному аэральному загрязнению НТО, будут пренебрежимо малы. Данные натурных экспериментов, учитывающие влияние реальных климатических факторов, могут быть использованы для тестирования региональных моделей воздушного переноса трития в системе «воздух – сельскохозяйственные растения».

Ключевые слова: *тритий, оксид трития, тритий в свободной воде, органически-связанный тритий, сельскохозяйственные культуры, аэральное поглощение, камерный и натурный эксперименты.*