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SOME OF THE SPECIAL FEATURES OF THE TRITIUM MIGRATION

Abstract. *It was found that tritium is different activity as part of the free water of plants can be conditioned by the processes of plant life, taking into account the weather conditions, composition of soil on which the plant is located. The paper shows the results of the field and laboratory studies on the search of the plants, which may act as a reliable indicator for determining tritium pollution of the environment. In this study, a plant that has a tritium high volume activity in the free water is considered an indicator of the environment tritium contamination, and / or organically bound tritium as a whole or in separate plant organs on comparable terms with other plants.*
Key words: *tritium, migration, isotope, environment.*

Introduction

The hydrogen is the most abundant element in the Universe. Taking into account the big penetration ability of tritium and possibility of easy exchange of atoms between the various isotopes of hydrogen, two main problems in the research of tritium may be named:

– The lack of the right understanding of transfer and bio-accumulation processes of tritium in natural surroundings and antropogenic environment [4, 13, 17, 18];

– The increasing need of the development of the new approaches to assessment of tritium human and nature in order to protect effectively [9, 11].

It should be noted that in Ukraine after the accident at the Chernobyl Nuclear Power Plant, work was actively carried out to investigate the migration of radionuclides in agricultural plants [24, 25, 31]. In these works the attention was alsooyven to tritium. These studies found further confirmation in works and experiments far beyond Ukraine [9, 11, 15, 34].

However, most of the studies dealed only with agricultural plants that grow on the territory that had been accurately exposed to technogenic tritium.

But due to the fact that tritium was not given proper attention for a long time. It often appeares a need to determine the contamination of a specific area with technogenic tritium. The question that appeares: is there a plant that can act as an indicator for the determination of tritium contamination of the environment?

The solution of this problem is relevant for monitoring of the condition of the zones that are around nuclear power plants and nuclear fusion enterprises, the repositories of radioactive materials that contain tritium. Another task is to assess the purity of recreational areas, agricultural lands and areas for grazing.

The results of the study, which are introducet below, were conducted in Ukraine in the territory of the enterprise, which until 2002 had been actively working with tritium. The work is based on the monitoring of the specific activity of tritium in birch sap, and the study of bioaccumulation of tritium in various herbaceous plants, that are common in the central part of Europe.

Partially published and unpublished results are presented in the this study.

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1. Tritium - the natural isotope of hydrogen

Tritium is a beta emitter and due to radiochemical properties it easily dissolves in liquids. Tritium is also easily absorbed by the human organism with water and promotes to damaging the health. Tritium is the most poorly known radioisotope [7, 30].

The chemical form of tritium (^3H) is a rare but it is natural isotope of hydrogen (H), and it is the only natural hydrogen isotope that is radioactive. from the perspective of atmospheric existence of tritium the most important forms. Are such forms as tritiated hydrogen gas (HT) and tritiated water (HTO). These tritiated forms chemically behave like hydrogen gas (H_2) and water (H_2O).

In 1957, the world first faced with a serious accident of environmental contamination by tritium. It was a Kyshtym disaster, a radioactive contamination accident, which occurred at Mayak, where a plutonium production center for nuclear weapons and nuclear fuel reprocessing plant in the Soviet Union was situated [30]. The accident was classified as a disaster of Level 6 according to the International Nuclear Event Scale (INES) [19], making it the third most serious nuclear accident ever recorded, following the Fukushima Daiichi nuclear disaster and the Chernobyl disaster (both Level 7 according to the INES). Now, the Kyshtym area is usually referred to as the East-Ural Radioactive Trace (EURT) [8].

And only now, the materials about the negative effects of tritium due to this accident become available to the general public. Residents of the villages that are situated around the scene of the accident have already been massively dying of cancer for a while. The majority of researcher's blame it on the usage of water with high specific activity of tritium. Pollution of rivers in the area of the accident has led to contamination of groundwater, and as a result plants and animals. The usage of products of plant and animal origin has become harmful to human organism [30].

In 1986 when the Chernobyl disaster occurred a lot of radioactive material precipitated onto much of the surface of the western USSR and Europe [29]. This accident was classified as an of 7 event (the maximum level of classification) according to International Nuclear Event Scale [3]. After the Chernobyl accident, approximately 14^{10} Bq of tritium got in to the environment. And 30 years after the nuclear accident look place, there is a high content of tritium in water, in the juice of birch, in products of plant and animal origin outside the exclusion zone [7]. In 2016 the New Safe Confinement, a structure that intended to contain dangerous remains of the unit No. 4, was built at the Chernobyl Nuclear Power Plant. But this New Safe Confinement does not protect from tritium which has already been in the environment. Particularly, at the 1-st International Conference on Nuclear Decommissioning and Environment Recovery INUDECO'16 was stated: "The Sarcophagus will protect the 4-th block of Chernobyl's NPP from the migration of radionuclides which are in the environment, that is not protect. Migration of radionuclides has not well been studied yet. The most poorly studied is tritium " [1].

In 2011 the nuclear disaster at the Fukushima-1 Nuclear Power Plant in Fukushima occurred. Japanese nuclear engineers have estimated that to bring the NPP into a stable and safe condition and liquidatie the consequences of accident they will need up to 40 years [27]. In January 2014, according to the report "Fukushima Nuclear Accident Update Log" [33], it was declared that a total of 875

TBq (2.45 g) of tritium are on the site of Fukushima Daiichi, and the amount of tritium that is contained in the contaminated water is increasing by approximately 230 TBq (0.64 g) per year [26].

The events that are taking pleace in the Eastern Ukraine (the anti-terrorist operation) have also gained relevance. Recently water contaminated by tritium penetrates from the war zone from coal mines in which conducted nuclear explosions in the USSR. It was also mentioned above that during the anti-terrorist operation the metallic components of various devices that contain tritium are applied. It may cause a global ecological disaster inside and outside of Ukraine [14].

Monitoring data indicate [12] that the technogenic component of tritium enters the environment in the following cases:

a) as a result of an operation of nuclear power plants, installations for nuclear fusion and emergency situations on these objects. The largest emissions of tritium accidents were recorded in Kyshtym accident, Chernobyl, Fukushima;

b) as a result of the disposal of tritium-contaminated products because of the humans of a person's technical work [7, 28]. Today it is the main source of penetration of technogenic tritium into the environment.

In January 2007, the Canadian Nuclear Safety Commission (CNSC) Tribunal directed CNSC staff to initiate research studies on tritium emissions in Canada [28]. Among there them are studies on the migration and bio-accumulation of tritium in plants. This is one of the fundamental researches in the designated sphere [22, 32].

There are many works on this subject of the French [5, 10, 16, 20], Chinese [11, 34] and Japanese [23] researchers.

But there are still many questions for the further research on the transfer of tritium in to the environment, the distribution and accumulation of radionuclides in plants [16, 21].

Since tritium irradiates the human organism from the inside, getting there with water and food, migration and bio-accumulation of tritium in the organic matter of plants is an actual topic of scientific research. As the Chernobyl and Fukushima accidents have shown, all the countries of the world are more or less affected by the consequences of such events [1].

2. Tritium in the environment

Hydrogen is one of the main substances required for the support of the vital activity and functioning of living organisms. The significance of hydrogen is caused by the exclusive significance of water in processes related to living organisms. Due to isotopic exchange, heavy hydrogen isotopes are able to participate in biochemical processes and to substitute hydrogen atoms with tritium atoms easy and readily.

The accumulation and migration of tritium in the environment depends on the time of occurrence and the location of tritium, the rate of exchange of tritium in air masses, and the concentration of tritium in the stratosphere during the exchange processes. In summer, the overground concentration of tritium at a height of up to 2 km increases due to evaporation of spring and winter atmospheric precipitations, and the oversea concentration of tritium at the same height decreases due to absorption of tritium by water. The circulation of atmospheric water due to the oceanic air exchange is very fast, so, as a result of a fast isotopic exchange between

water drops and water vapors, rains transfer tritium to the lower layers of the troposphere [30].

In the process of radioactive decay of tritium, beta particles and $\overline{\nu}_e$ antineutrinos are emitted. The reaction of radioactive decay of tritium can be described as following:

$$^{3}_{1}H \rightarrow {}^{3}_{2}He + e^{-} + \overline{\nu}_e .$$

The half-life period of tritium is 12.32 ± 0.02 years. The average energy of tritium beta particles is $E_\beta = 5.52 \ \ldots \ 5.7 \ keV$. Tritium beta particles expend the basic part of their energy on interaction with the electron shells of atoms of substances that are in the environment where the beta particles move. The isotopic composition of hydrogen and oxygen, which goes from water to plant hydrocarbons in the photosynthesis process, depends virtually completely on the isotopic composition of water. Hydrogen isotopes fractionate during processes of water evaporation and condensation. Different ratios of $^{18}O/^{16}O$ and $^{3}H/^{2}H/^{1}H$ depend directly on the average annual temperature. The water condensation temperature has a significant effect on the isotopic composition of water, that hos been confirmed by the results of some studies.

The transformation of aqueous tritium (NTO) into organically bound ^{3}H isotope depends on the selectivity of ^{3}H isotope that is relative to ^{1}H isotope. As a result the penetrotion of ^{3}H isotope in organic compounds is reduced by approximately 20 percent. It means that the ratio of the maximum specific activity of ^{3}H isotope in organic fractions to the maximum specific activity of ^{3}H isotope in free plant water is about 0.05 within several days after a single ingress of the isotope. It is assumed that the specific activity of ^{3}H isotope in free water and the specific activity of organically bound ^{3}H isotope increase exponentially with time, in case that ^{3}H isotope enters the environment continuously [7]. But the studies and observations, which are mentioned below, have demonstrated the polynomial time dependence of these processes.

The studies that are performed were based on the methods developed in the seventies of the 20th century [2]. The studies for the determination the interaction of tritium with tree tissues were commenced approximately in the middle of the 20th century, and the studies for determination of the distribution of tritium in the tree annual growth rings were commenced in the eighties of the 20th century. It was detected that before the 1990s the activity of tritium in rare atmospheric precipitations was lower than the activity of tritium in tree tissues due to the accumulation of tritium in tree tissues. After nuclear tests were finished, the concentration of tritium in the atmosphere began to decrease gradually. The process of the removal of tritium from trees gets slower due to the accumulation of exchangeable tritium. These conclusions are confirmed by the results of studies mentioned below.

In the ground, tritium exists in two basic forms. Then main part of tritium is in free ground water (NTO) and corresponds to the concentration of tritium in atmospheric water. The upper 4 cm of ground layer contains up to 25 percent of all tritiated water. When the depth of the ground layer increases, the content of tritiated water decreases up to 4 percent in the 14 cm to 16 cm ground layer, and

then increases to 10 percent in the 16 cm to 18 cm ground layer. Below 16 cm, the content of tritium is 40 percent of the total content of tritium in ground [7]. After removal of free ground water, some part of tritium remains in the ground in ion-exchangeable and sorbed forms.

That is, tritium, as a hydrogen nuclide, is characterized by its high migration capability in water and, as a result, by its high activity in the exchange processes in the living organisms and by its mobility in water-plant systems.

Roots and other underground organs of plants are relatively intensively enriched with tritium. The distribution of organically-bound tritium in plant tissues and organs is nonuniform and depends on the biochemical properties of plant tissues and organs. Plants and ground bacteria are the catalysts of oxidation of gaseous tritium (NT) that is contained in the atmosphere. The rate of transformation of gaseous tritium into aqueous tritium in the ground vegetation is about 1 percent per 48 hours. Because of this, the specific activity of tritium in ground moisture increases as compared with the specific activity of tritium in free plant water [7]. But the results of studies performed within 10 years indicate that these statements are not applicable for all plants.

3. The study of the air-water-plant system

As a base statement in studying on the migration of tritium over the 15-year period, it was accepted that the specific activity of tritium ^{3}H in free plant water can be the same in all of organs plant only if the activity of tritium is the same in atmospheric moisture and ground moisture, otherwise there are essential differences in the specific tritium activity values, which are caused by the tritium concentration gradient in the air-water-plant system [7]. But the results of some additional measurements, which were performed with consideration of meteorological conditions during sampling and averaged through the month, provide the possibility to state that the high concentration of tritium in free plant water can be caused by the processes of vital activity of plants, with the consideration of temperature, humidity, and composition of the ground where the plants being studied that cae located.

The obtained results provide the possibility to determine the total contamination of the territory with tritium and the effect of contamination of plants in the territory. The results also provide the possibility to study the process of biological accumulation of tritium in plants, specifically in Bétula péndula, due to water exchange. Figure 2 shows the characteristics that are illustrate the variations of the specific concentration of tritium in meltwater and birch sap that are obtained during the 13-year period of studies.

As is seen from the characteristics, if tritium continuously enters the environment, the specific activity of tritium in free water and the specific activity of organically bound tritium varies with time according to a polynomial expression instead of an exponential expression.

On the characteristic basis, there is the drastic increase of the specific activity of tritium in birch sap in 2008, when the specific activity of tritium in snow cover melt water significantly decreased. In the next years, the specific tritium activity values vary according to regularity, that is, the specific activity of tritium in birch sap increases in the following year after the increase of the specific activity of tritium in meltwater that was detected in the preceding year.

The specific activity of tritium varies according to the 5-order polynomial expressions showed below.

The specific activity of tritium in meltwater:

$$y = 0.0553x^5 + 2.2468x^4 - 33.043x^3 + 213.31x^2 - 608.23x + 956.05.$$

The approximation coefficient $RI = 0.5$.

The specific activity of tritium in birch sap:

$$y = 0.0444x^5 + 1.8959x^4 - 28.135x^3 + 163.42x^2 - 280.1x + 270.26.$$

The approximation coefficient $RI = 0.3211$.

The low values of the approximation coefficient are caused by the significant changes of the specific activity of tritium in meltwater in the range from an absolute minimum to an absolute maximum. This feature allows to make certain conclusions.

In the preceding years, the researchers in Ukraine [7] that performed studies in contaminated areas, such as the Chornobyl area and territories with radioactive waste storages, stated that elimination of tritium from plants is possible due to water exchange. The basic half-life period, which is typical for the elimination of 90 percent of aqueous tritium, virtually does not depend on climatic conditions and ranges from several hours to (10–20) days.

The characteristics in Figure 2 show that the part of tritium that was detected in meltwater in February, and which, according to the aforesaid statement, should cause the increase of concentration of tritium in birch sap in March and April, is not included in the water exchange process. The next year, the specific concentration of tritium in birch sap sometimes significantly increases, while the concentration of tritium in meltwater decreases.

These facts can be explained by following.

Firstly, in the years when the concentration of tritium in birch sap was increased, the air temperature and humidity during the study period were increased. That is, this period was characterized by more intensive birch sap circulation in favorable for vegetation conditions.

Secondly, tritium in meltwater that enter clayed ground remains in the ground. The tritium atoms participated in the atomic exchange with hydrogen atoms in some clay-containing minerals and free ground water. The temperature fluctuations decelerated (when the tritium concentration decreased) or accelerated (when the tritium concentration increased) this exchange was a normal chemical reaction in the presence of a hydrogen atom. Because of this, part of tritium in was meltwater was accumulated in the ground.

Thirdly, the aforesaid data indicate the significant increase of the concentration of tritium in the environment at the end of winter and at the beginning of spring, that is, during the periods when processes of intensive evaporation and humidification on the ground surface are intensive. The polynomial expressions for changes in the specific activity of tritium in ground water and tritium in birch sap demonstrate the cyclic decrease of the specific tritium activity, in the range from the minimum activity to the maximum activity,

at least four times within a year. According to the polynomial expressions the cycles correspond to rainy periods. For this reason, it is possible to assume that some plants that grow in the territories contaminated with tritium can accumulate tritium in the plant organs or some tissues during the plant vegetation period. The period of accumulation of tritium in plants and the period of elimination of tritium from plants depend on the temperature of the environment, air humidity, and, probably, chemical composition of the ground where the plants grow.

The question that emerges is the following:

How tritium should migrate and accumulate insomuch that the specific activity of tritium in birch sap will increase the next year?

4. The field and laboratory experiment

In order to confirm the aforesaid assumption, there was carried out a field experiment. In the period from May till October 2016, aqueous extracts from about 100 plant species were studied for determin action of the specific activity of tritium in different periods of the plant life cycles.

The study results provide the possibility to state that there are plants which accumulate tritium in the plant organic substances for some period of time. It was experimentally determined that the specific activity $\boldsymbol{A}$ of tritium in the extracts of common taráxacum (Taraxacum officinale Wigg.) was higher as compared to the extracts of other plants, provided that the plant samples were taken from the plants that are located at a distance of no more than 1 m from each other.

The results of one of these samples from May 28, 2016, are shown in Table 1.

Table 1. Measurements of the specific activity of tritium in plant extracts

#	Place of collection	Plant	$\boldsymbol{A}$, Bq/l	Part of the plant
1	Zone (A)	Bétula péndula	10152.5	Green seed
2	Zone (A)	Trifolium	12673.8	Flowers
3	Zone (A)	Plantágo	11606.8	Leaf
4	Zone (A)	Taráxacum	38444.4	Stem and post-flowering flower
5	Zone (B)	Taráxacum	7967.24	Leaf

Zone (A) is a section with a radius of 300 m (sanitary zone), which is directly adjacent to one of the corps of the company working with tritium. Zone (B) is located at a distance of more than 500 m from the production building of the mentioned enterprise.

The ambient temperature during sampling was within + $(20\text{-}26)^0$C, the relative humidity of the air fluctuate within (76-84)%.

200 measurements of aquatic extracts of taraxacum were investigated in different parts of the study area. The results allowed to assert that the specific activity of the aquatic extract of the taraxacum is always higher in comparison with the extracts of other plants that grew along with the sample of the taraxacum.

It can be noted that within the sanitary zone, the specific activity of the water extracts of Taraxacum officinale Wigg. has much higher indicators than analogical measurements in a relatively clean zone. It should be noted that while the aquatic extracts of other plants from these sampling points did not have similar high rates. Results of measurements of extracts of other plants accounted for (10-46)% of the indicated values.

There were made a number of measurements. These measurements confirmed the increased content of tritium in the extract of taraxacum. Subsequently, the systematization of measurements was made and the hypothesis was accepted: in different parts of taraxacum, the content of tritium is different. To test this hypothesis, several plants were selected from the Zone (A) of the previous study. Taraxacum from Zone (B) was taken from a distance of 150 m from the boundary of Zone (A). These plants ware intact, almost the same size and weight.

From the roots, leaves, stems and flowers separately, weighing 2 grams, extracts were made and their specific activity was checked. The results are shown in Table 2.

Table 2. Measurements of specific activity of tritium A in the extract different parts of Taraxacum officinale Wigg.

#	Place of collection	A, kBq/l			
		Root	Leaf	Stem	Flowers
1	Zone (A)	10.90	47.29	50.60	24.44
2	Zone (A)	13.41	16.29	14.60	18.13
3	Zone (A)	14.07	18.76	14.93	28.67
4	Zone (A)	11.42	16.13	27.59	17.07
5	Zone (B)	10.27	11.98	14.51	15.78

Plants 2 and 3 grew at the same place (two plants next to each other). They were almost the same in size and weight of the plant, but for to one difference: the plant 2 was already blossoming, and 3 - still in the bud. Indicators of the root, leaf and flower pedicle had no significant differences. Plants 5 had a flower that was blossoming, but not dried yet.

Further studies of tritium in Taraxacum officinale Wigg. occurred in June. Weather conditions: an increase of air temperature and a decrease of moisture. In addition, the taraxacum has already ended the growing season. All available plants from the study area did not have flowers.

In the course of further measurements it was proved: with increasing heat, the specific activity of tritium in plant extracts is reduced. At the same time, the amount of latex of taraxacum decreases and the plants become rough, the flower turns into seeds, the stalk dries. Increasing air temperature and the transition of Taraxacum officinale Wigg. to "summer calm" the content of tritium in organic matter of plants decreases. Experiment with water that was obtained from transpiration of plants showed that the concentration of tritium in transpiration water and in the soil does not increase during transpiration period (7 days). Transpiration water was obtained by installing a glass cube with collectors over the plant. The walls of this device were deepened into the ground and sprinkled with some soil, to limit the flow of moisture and air from the outside.

After establishment a dry weather with a temperature above + 27^0C, the specific activity of tritium in aqueous extracts of taraxacum from the all over study area has become approximately the same.

To confirm the uneven distribution of tritium in the free water of particular plant organs of Taraxacum officinale Wigg. there was carried out a laboratory experiment.

In a separate container with soil that is similar to the soil of the study area, were planted Taraxacum officinale Wigg. and random plants, that were typical for the study area. The first container – is the main one, the second container is used to confirm the results. Each container was placed in a single greenhouse.

Plants were watered tritional's water once. Further watering of the plants was only with water. The humidity in the containers was maintained at (76-86)%, air temperature was + $(22\text{-}26)^0$C. Observations of the plants took place over three weeks, with recording of external changes during the growing season (emergence of a bud, blooming flower, number of new buds, growth rate).

After two weeks ofter the start of the experiment, when the plant threw out the first bud and started to grow rapidly (stem grew during the day up to 5 cm), samples were taken of the specific activity in extracts of particular parts of plants (at 1 gram), as well as similar samples from other plants in container – Persicaria lapathifolia L. (tab. 3). In the same period in the environment, to the extent not included in the sanitary zone the same (natural) taraxacum were taken for the study of the specific activity of tritium in the composition of the free water of plants (table. 3).

Table 3. The results of laboratory experiment

Part of a plant	A, Bq/l			
	Laboratory conditions		Natural conditions	
	Taraxacum officinale Wigg.	Persicaria lapathifolia L.	Taraxacum officinale Wigg. # 1	Taraxacum officinale Wigg. # 2
Flower	3849.89	1530.63	195.17	92.73
Stem	9923.06	1509.55	230.06	101.74
Leaf	6109.85	1522.98	156.03	64.84
Root	2288.04	1768.53	138.37	42.91

Figure 1 shows the comparative chart of the accumulation of tritium in particular parts of the plants.

A similar distribution of the concentration of tritium is observed in taraxacum # 1 and # 2.

Thus, considering experimental data, it can be argued that the presence of tritium in the air, in the soil and/or groundwater, Taraxacum officinale Wigg. will accumulate a large part of the tritium in the stem and partially in leaves and flower during the growing season.

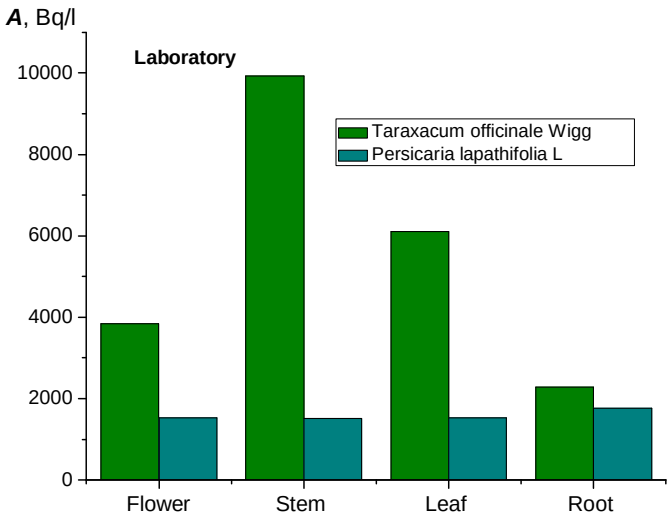

Fig. 1. Diagram of the tritium activity in particular parts of plants.

Considering the results, we can deduce the primary mechanism of tritium migration in Taraxacum officinale Wigg.

Conclusions

One of the main objectives of these studies was to quantify tritium migration and bioaccumulation in organic substances of plants.

This paper contains the data on the preliminary results of the studies and the data on the primary processes promoting tritium migration, which were obtained for common taráxacum plants.

The obtained results are the following:

1) If tritium enters the environment continuously, the concentration of tritium in free water and the concentration of organically bound tritium increase with time according to a polynomial expression.

2) The base statement was confirmed. The concentration of tritium in free water of plants, as assumed, is virtually the same in all plant organs only if the tritium concentration in atmospheric moisture and ground moisture is the same, otherwise there are essential differences in the specific tritium activity values due to the gradient of tritium concentration in the atmosphere-plant-ground system.

3) It was detected that the different concentrations of tritium in free plant water were caused by processes of vital activity plant of depending on air temperature and humidity.

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