



## Conference Paper

# Radiological Problems of Tritium

**B. I. Synzynys, O. A. Momot, O. A. Mirzeabasov, A. V. Zemnova,  
E. R. Lyapunova, Yu. M. Glushkov, and A. A. Oudalova**

Obninsk Institute for Nuclear Power Engineering of the National Research Nuclear University MEPhI, Studgorodok 1, Obninsk, Kaluga region, 249040, Russia

## Abstract

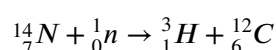
In light of the session documents (UNSCEAR, 2015, 2016), presented are the results of health risk assessment in drinking water containing tritium as well as results of field observations and bioassay with algae (*Lemna minor* and *Polyrhiza*) from water bodies near radiation-dangerous industrial and scientific objects. The prospects for future studies in these areas should be associated with the assessed human radiation dose from organically bound tritium in organs and tissues of people, animals and plants; the search for plants and animals selectively accumulating tritium and its subsequent bioassay in water. It is necessary to develop the approaches to harmonization of tritium standards in water objects and potable water, in particular. Sanitary and hygiene standards are suggested to develop on the basis of health risk assessment in drinking water with tritium. The upcoming trend for assessing the contribution of organically bound tritium to the total human radiation dose is the local irradiation by tritium incorporated into DNA of cells and tissues and radiosensitive organs. The well-known and new methods of microdosimetry of DNA incorporated radionuclides are necessary to solve this problem.

**Keywords:** natural and technogenic tritium, water biomonitoring, *Lemna minor*, *Spirodela polyrhiza*, health risk assessment, drinking water, the interventional level for tritium.

## 1. Introduction. Tritium sources on the Earth

Tritium (symbol  $^3\text{H}$  or T) is  $\beta$ -radioactive hydrogen isotope with a half-life of 12.35 year. The maximum energy of  $\beta$ -particles is 18.6 keV, the average energy is 5.7 keV. In the Earth's biosphere tritium is balanced with the stable hydrogen isotopes: protium  $^1\text{H}$  and deuterium  $^2\text{H}$  (or D) in groundwater, gaseous fluids and living matter.

*Natural tritium.* The natural level of tritium is one in  $10^{18}$  protium atoms. It is the result of nuclear reactions under the action of space rays in the upper atmospheric layers.



Corresponding Author:

B. I. Synzynys  
synzynys@obninsk.ru

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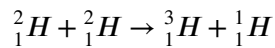
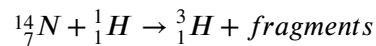
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After thermonuclear weapon testing (1954) the atmospheric concentration of tritium had increased a hundredfold, however, by now it lowered as a result of prohibition against these tests and natural decay. In recent years the nuclear power plants which annually release several tens of kilograms have become the primary source of technogenic tritium in the environment.

### 1.1. Tritium generation in NPP operation

At NPPs tritium is formed in the reactors as a product of ternary fission; as a result of (n,  $\gamma$ ) deuterium nuclei reaction in the coolant, i.e. water; in the neutron capture by boron and lithium nuclei in the coolant, i.e. water (in boron regulation and water regime correction – at NPPs with WWR) and in control rods; as a result of  ${}^3\text{He}$  (n, p)  ${}^3\text{H}$  reaction in a gas filling the graphite stack at NPPs with high-power pressure-tube reactors; as a result of (n,  ${}^3\text{H}$ ) and (n, p) fast-neutron reactions in  ${}^{14}\text{N}$ ,  ${}^6\text{Li}$ ,  ${}^{10}\text{B}$ ,  ${}^{40}\text{Ca}$  and other nuclei available in different materials applied in the reactor structure. Some reactions of tritium formation proceed just in reactor water and partly in fuel elements and control rods. Tritium from fuel elements and control rods enters the reactor water in the loss of their tightness and due to cladding diffusion or escape because of leaky cladding.

At NPPs with pressurized water reactors having boron regulation, the primary reaction of tritium formation in a coolant is the reaction  ${}^{10}\text{B}$  (n,  $2\alpha$ )  ${}^3\text{H}$ , in the absence of boron regulation it is the reaction of deuterium neutron activation, i.e.  ${}^2\text{H}$  (n,  $\gamma$ )  ${}^3\text{H}$ . Tritium formed in a coolant is supplemented by tritium from fuel elements and control rods.

The power of tritium sources is of about  $5 \cdot 10^7$  Bq/day in a unit at most. Natural tritium entering NPPs with the fresh air ventilation is also included into these values. The assessed atmospheric emission of tritium by NPPs is accepted not more than several units of  $10^8$  Bq/day. It produces the volume activity of tritium in the surface atmospheric layer which is equal to several units or several tens of units Bq/m<sup>3</sup>. At this volume activity of tritium in the surface atmosphere the radiation exposure of individuals will be not exceed 10 mSv/y.

Direct measurements of tritium content in the surface atmospheric layer near the Chernobyl NPP before the accident 1986 were of about 0.5 Bq/m<sup>3</sup>, at the Ignalina NPPs up to 1 Bq/m<sup>3</sup>. It means that the radiation exposure in tritium inhalation is 10

times less than the above value and, hence, does not exceed the permissible dose for people after NPP radioactive discharge. In 1992 at the Kalinin NPP the volume activity of tritium in the surface atmosphere was  $0.1 \text{ Bq/m}^3$ . The presented values show why atmospheric emissions of tritium are not normalized and why there is no need to control the environmental discharge by NPPs.

Most of tritium «worked» at NPPs falls away with liquid effluents. Liquid effluents by many NPPs are discharged into heat-sinks and tritium found after the leakage enters these heat-sinks too, i.e. technical (used for equipment cooling), unbalanced and other types of water removed from NPPs. It can be believed that water in the primary circuit or multiple forced circulation circuit will be renewed during one year NPP operation and, hence, the whole worked tritium and that found in these circuits will enter the water-cooling pond (except tritium discharged into the atmosphere, its fraction is below 20%).

Tritium is the unique radionuclide. Being actively introduced into living organisms, it can violate the structure of biologically important molecules in cells not only via internal beta-radiation, but also as a result of the following transmutation into  $^3\text{He}$ :  $^3_1\text{H} \rightarrow ^3_2\text{He} + e^- + \bar{\nu}$ . Antineutrino extricated in the reaction is suggested to be biologically inactive.

The excretion half-life of tritium from an adult organism is approximately 11 days. Tritium beta-particles (the mean free path in water is  $0.5 \mu\text{m}$ , and the maximum one is  $6 \mu\text{m}$ ) do not penetrate through epidermis. If, however, tritium falls within an organism, and the more so, into cells, its radiation can irradiate the inner parts of a human body. It should be appreciated that the linear ionization density by tritium beta-particles is higher than in case of alternative radiators.

Tritium is a part of tritiated water HTO in oxygen interaction and as a result of isotopic exchange with protium in water molecules. Its chemical properties are similar to ordinary water. Under natural conditions water with the specific activity of tritium  $1 \text{ Bq/l}$  contains about one tritium atom in  $10^{18}$  molecules of ordinary water and the “natural” content of tritium in fresh underground and surface water in Russia is 2–6 Bq/l.

**The aim** of this research is to review the proper studies of distribution, biological impact and radiation health risk assessment of natural and technogenic tritium in light of new UNSCEAR documents.

## 2. Organic substances with tritium

Tritium is well exchanging with other hydrogen atoms, for this reason a certain fraction of tritium absorbed by animal and plant tissues is included into organic compounds of the fat, protein, nucleic acid and collagen type. This tritium is specified as organically bound tritium (OBT). It should be noted that tritium atoms in OBT, being chemically combined with carbon atoms, are effectively fixed in different biomolecules until the compound begins metabolizing, i.e. it is practically unable to exchange in the presence of biosynthesis or, vice versa, decay. However, tritium atoms fixed with oxygen, sulphur, nitrogen or phosphorus atoms are ready to exchange with water hydrogen and are not considered as OBT or classified as exchangeable OBT [1]. OBT has a long lifetime, i.e. 40 days and more in a human body. It is also right even for OBT classified as easily exchangeable.

At present a wide variety is used of tritium labeled organic substances including biochemical ones for investigations. The personnel dealing with these substances may be exposed to radiation in inhalation, skin contact or accidental ingestion. Labeled biochemical substrates (amino acids, DNA precursors, glucose, hormones) can metabolize in a human body and partly lose the tritium label which is included in other macromolecules, being turned into OBT [2, 3]. Labeled DNA precursors such as H<sub>3</sub>-thymidine and H<sub>3</sub>-cytosine deoxyribose are also widely used in scientific research. In mammal organisms they are partly degraded into HTO and included in DNA of dividing cells, from this moment on they locally irradiate the cell nuclei by electrons. It is the most dangerous type of irradiation, as the whole energy of radiation is released in genes and transmutation of tritium into helium-3 takes place; this process disarranges the DNA structure too [4].

Slightly soluble tritium compounds (luminous powders, metal tritides applied in proton accelerators to have neutron currents of a given energy and density) are widely produced and utilized in industry and research. Tritium found in the water bodies near Obninsk probably "comes out" of these compounds.

All nuclear states are active in tritium application, however, the regulatory system for safe operation has not been developed yet.

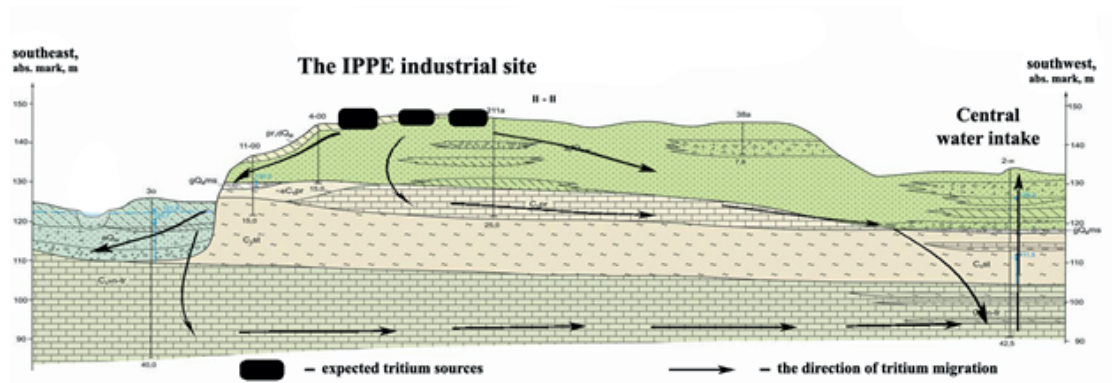
### 3. On the need for harmonization of tritium standards in the international and national radiation safety standards

As noted above, the official UNSCAR documents [5, 6] state that «for some reason the problems associated with tritium contamination of water objects are of special interest for radiobiologists, radio ecologists and radiologists». It is due to the fact that the radiation-hygienic standards on tritium are much different in the countries with “radiation legislation”. The sanitary legal system of various states declares such a standardized index as “the intervention level” of tritium content in portable water, Bq/l: Ukraine – 30 000; Republic of Belarus – 7 700; Republic of Kazakhstan – 7 700; Russian Federation – 7 600; Canada – 7 000; the USA – 740; EU countries – 100. Such a significant difference is supposed to be caused by several reasons: incompleteness of the sanitary-hygienic legislation that results from insufficient studies of radiological effects of tritium and, ultimately, the problem of OBT pharmacokinetics is not brought to light; the instrumental base for tritium detection is imperfect; the models for assessing the internal exposure dose by tritium oxide and, the more so, by OBT have not been developed completely [6-8].

The methodology of radiation risk assessment is proposed to be a common platform for normalization of radiological tritium standards [10].

### 4. Health risk assessment in drinking water contaminated with tritium

The primary problem at present is to assess the human body tritium intake and to determine the role of tritium incorporation into the organic component of tissues [7]. L.A. Chipiga and M.I. Balonov have analyzed 9 papers published in 1968-1997; 3 chamber models of tritium distribution for its ingress into the human body in a form of tritium water, including MKPZ, have been used. It is noted that according to the MKPZ model, the nominal OBT fraction from tritium water intake is 3%, and according to other models it is 0,4 и 0,8%, respectively. This made possible the estimation of OBT contribution to the average body radiation dose: according to the first model it is 3%, and for two other ones 3,6 and 9%, respectively, of the total effective dose formed in the organism as a result of tritium ingress. It must be emphasized that radiation of OBT incorporated into DNA cells, protoplasm or intercellular fluid will result in different biological effects.

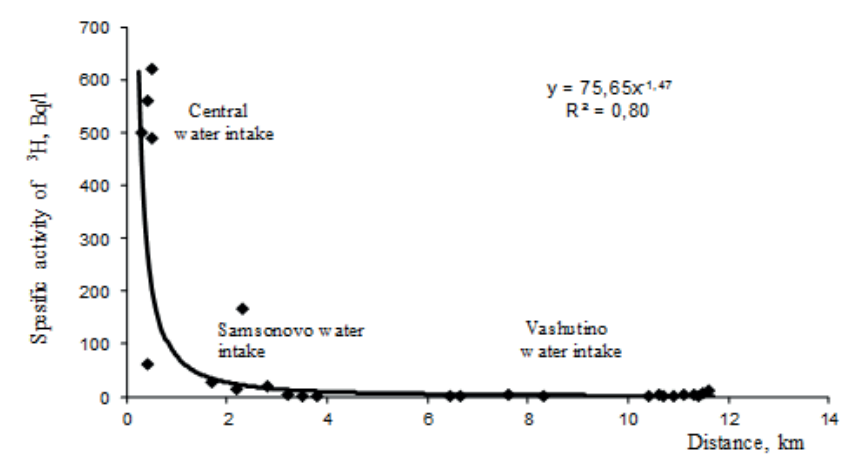


**Figure 1:** The scheme of tritium migration from the radioactive waste storage into the river Protva and groundwater. The Central water supply intake of Obninsk (to the right) [12].

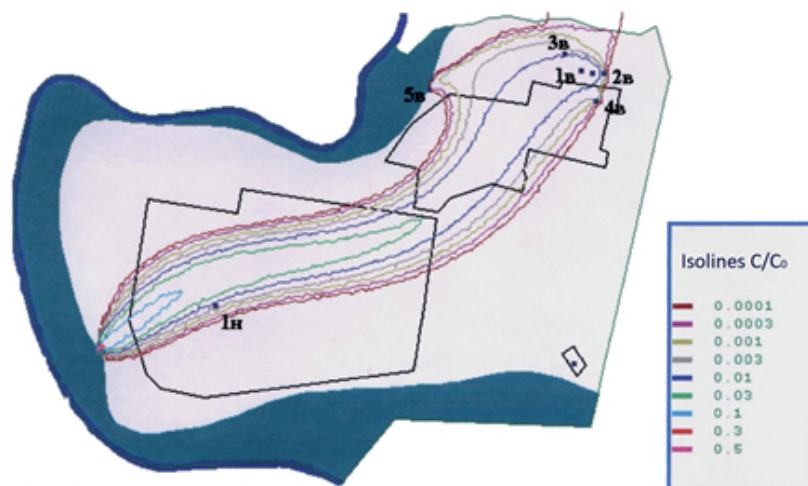
Let's consider groundwater tritium pollution in Obninsk and estimate the radiation risk by a linear nonthreshold theory of low radiation effects. Groundwater tritium pollution was first detected in the RF SRC – IPPE sanitary protection zone (Obninsk) by Roshydromet employees in 1997 within the «Radiological monitoring program of nuclear power and industrial facilities» [11]. Detailed studies of the IPPE industrial site and the adjoining territory had shown that the sources of technogenic tritium were nuclear reactors, accelerators with tritium targets as well as radioactive waste storages. Measurements of the specific tritium activity in the river Protva oxbow near the RF SRC – IPPE have shown the following. In 2005 the specific activity of tritium ranged from 6290 to 37000 Bq/l, in 2010 from 730 to 1830 Bq/l, in 2017 from 550 to 3600 Bq/l [12, 13]. The radioactive waste storage could be the source of tritium contamination of the river Protva, its oxbow and groundwater. Tritium, by virtue of migration capacity, had crossed the protective barriers and was found in the environment. Figure 1 shows the scheme of expected tritium distribution from the radioactive waste storage into the river Protva (to the left) and groundwater.

As Figure 1 shows, water with tritium from this storage, when falling into r.Protva and groundwater, feeds the Central supply intake. Therefore it is no wonder that in 2005 the enhancing tritium content was detected in the Central supply intake. Tritium concentration in water of other supply intakes was much lower. Tritium concentrations for all three water supply intakes are presented in Figure 2.

As seen, the specific activity of tritium in the water intake which is the nearest to the Central one exceeds 600 Bq/l. In Samsonovo intake it is about 50 Bq/l and technogenic tritium is practically absent in Vashutino water intake.



**Figure 2:** Tritium specific activity in water of Obninsk supply intakes depending on the distance from the Central water intake.

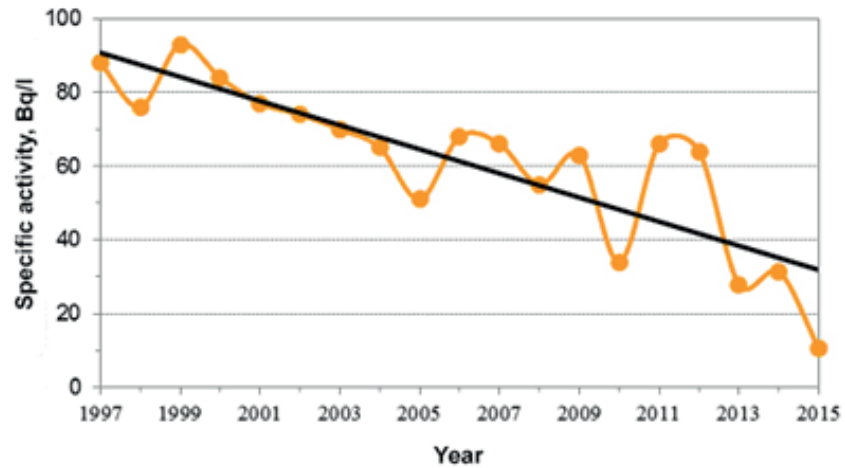


**Figure 3:** Propagation direction of the groundwater "tongue" with tritium oxide.

Early in the 2000s Obninsk geoscientists have determined the direction of the primary tritium stream in groundwater (Figure 3); it moves north-eastward going round Balabanovo and Borovsk and running to the southeast outskirts of Moscow.

These data give evidence of available sources of tritium in the territory of Obninsk and its ingress in groundwater. It can be also concluded that there is the tendency for its content reduction in water objects due to radiation decay as well as to the decrease in source capacity.

The individual life-long risk for Obninsk inhabitants has been estimated under the condition of permanent drinking of tap water with tritium. Figure 4 presents the yearly



**Figure 4:** Dynamics of tritium concentration in drinking water within the SPA "Typhoon" territory [14].

dependence of specific tritium concentration in Obninsk tap water within the SPA "Typhoon" territory.

According to the nonthreshold theory of a radiation dose dependence of stochastic effects, the risk  $R$  (1/year) is proportional to the effective irradiation dose  $E$  (Sievert). In line with RSS 99/2009 [15], the value of  $R$  is related to the dose via linear coefficients  $r_E$  of radiation risk by

$$R = r_E \cdot E. \tag{1}$$

Data on the average tritium content (10.6 Bq/l) in drinking tap water for 2015 have been used in calculating. The absorbed dose  $D$  (rad) of tritium beta-radiation was assessed from the equation (2) [16]:

$$D_{\beta}(t) = 2,0 \cdot 10^{-3} \cdot \frac{\bar{E}_{\beta} \cdot A_v \cdot V \cdot f \cdot T_{\text{eff}}}{m} \cdot \left( t + \frac{T_{\text{eff}}}{0,693} \cdot e^{-\frac{0,693 \cdot t}{T_{\text{eff}}}} - \frac{T_{\text{eff}}}{0,693} \right), \tag{2}$$

where  $t$  – exposure time, day;

$\bar{E}_{\beta}$  – average energy of  $\beta$ -radiation decay, MeV;

$A_v$  – specific activity of tritium in drinking water, Bq/l;

$V$  – intake rate of drinking water, l/day;

$f$  – transfer coefficient in the critical organ (1);

$T_{\text{eff}}$  – the effective half-life, s;

$m$  – body weight, g.

The absorbed tritium radiation dose was calculated on the hypothesis of equilibrium distribution of tritium oxide round the human body of 70 kg in mass and in case of drinking 2 l/day of tritiated water during a year. According to our calculations, the



absorbed radiation dose is  $2 \cdot 10^{-7}$  Gy, and it is much lower than the annual maximum permissible dose (RSS 99/2009 [15]) of 1 mSv for population. The relative biological efficiency (RBE) for beta-radiation of tritium incorporated into living organisms [17, 18] for carcinogenic effects was chosen as 2.5. The effective whole-body dose of  $E = 3 \cdot 10^{-7}$  Sv was calculated from these data. The risk of malignant disease in drinking water with trace amounts of tritium is assessed as  $1,6 \cdot 10^{-8}$ . The obtained health risk assessment value is 3 000 times less than the admissible individual life-long risk for population. Therefore it may be concluded that malignant tumors of Obninsk inhabitants may be caused by different reasons but not tritium effects.

## 5. Bioassay of natural water contaminated with tritium

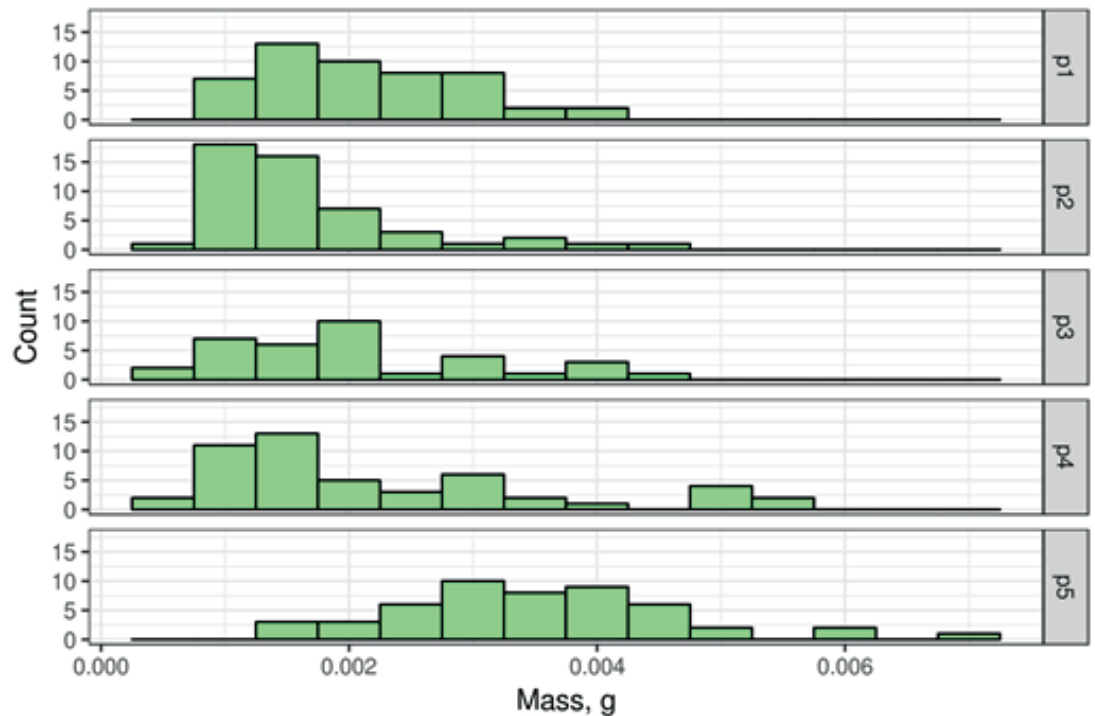
This research is aimed at studying the morphometric features of *Lemna minor* and allied species *Spirodela polyrhiza* to develop the radiation (or combined chemical and radiation) bioassay for screening surface and groundwater contaminated with tritium compounds. In future the data will allow interpreting the results of ecological and health risk assessment in drinking water with trace amounts of tritium.

To assess the specific activity of tritium, water and algae samples were taken from the oxbow of r. Protva and just the river in the SRC- IPPE affected zone.

After special sample preparation the specific activity of tritium in water was determined by a scintillation spectrometer Quantulus-1220 using internal standards and reference water samples. It is found that the specific activity of tritium in different Protva river oxbow areas ranges from 550 up to 3693 Bq/l; on July 6, 2017 the specific activity in the nearest Protva river oxbow area was 21.5 Bq/l.

Morphometric features of *Lemna minor* and allied species *Spirodela polyrhiza* were determined by a special camera microscope; the statistical data processing has been realized with the information space R software [19]. 100 algae and polyrhiza plants were used for every sampling "site". Distribution functions were plotted from the corresponding parameter values, for which estimated were the indices corresponding to median values, the first and the third quartiles of distribution functions. These data presented in Figure 5.

The upper plot, site 1 (right symbols) is the mass distribution of *Lemna minor* fronds dwelling in a water body with the specific activity of tritium of 650 Bq/l. The bottom plot, site 2 is the mass distribution for a water body with tritium activity of 654 Bq/l. Much the same activity was in the third water basin. Algae plants dwelling in the river Protva mainstream at site 4 with the specific activity of tritium 21.5 Bq/l were taken as



**Figure 5:** Mass distribution of *Lemna minor* fronds depending on tritium concentration in river water.

the reference ones. It is of interest that in a water body with the highest activity the algae mass distribution is right shifted, i.e. high tritium activity stimulates these plants growth.

*Spirodela polyrhiza* plants do not respond to tritium radioactivity in the concentrations found and evidence their relatively high radiation resistance. Quite the opposite, algae plants demonstrate the stimulated growth and development at higher values of tritium radioactivity.

## 6. Conclusion

The complex problem of tritium is considered not only from the technical (sources, elevated tritium concentrations as compared to the background values) but also biological (its impact on biota and health risk) viewpoints; the importance of determining OBT to assess the risk of low tritium doses and their effect on human organism has also been discussed.

The assessed radiation risk in drinking water contaminated with tritium in concentrations of 10.6 Bq/l gives evidence of unavailable extra cases of stochastic irradiation effects for Obninsk inhabitants.

Bioassay has shown that *Lemna minor* plants are sensitive to high activity of tritium in natural water. In this case their sensitivity manifests itself in stimulating the growth and development of aquatic plants at tritium concentration (activity) of 3600 Bq/l in the water bodies of Obninsk. The morphometric and other features of *Lemna minor* and *Spirodela polyrhiza* are to be studied under long-lasting effects of higher tritium concentrations as compared to the intervention level declared by RSS 99/2009 [15].

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