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Tritium in the Environment of Gulf of Finnland

M.A. Kulkova, A.V.Davidochkina

***Abstract*—Tritium is a radioactive form of hydrogen, used in research, fusion reactors and neutron generators. The form of most concern, tritium oxide (HTO), is generally indistinguishable from normal water and can move rapidly through the environment in the same manner as water. The expanding construction of nuclear industrial plants and nuclear power stations on the shores of the Baltic Sea (Gulf of Finnland) is creating a real possibility for the introduction of radioactive wastes into the vegetation and the waters of Baltic Sea basin (Sea water, Ladoga Lake, St.Petersburg rivers). A low-level liquid scintillation system Quantulus 1220 (Wallac, Turku, Finland) and system for sample preparation Sample Oxidizer 307 (PerkinElmer) were used for measurements of vegetation and water samples from this region. Significant difference was observed on the distribution of tritium concentrations in different types of water, snow cover and vegetation of Finnish Bay basin.**

***Index Terms*—Baltic Sea basin, Nuclear Power Plants Quantulus 1220, tritium pollution.**

I. Introduction

Tritium is a radioactive isotope of hydrogen. Tritium is produced naturally by cosmic rays but is also a by-product of nuclear power generation systems which use deuterium or "heavy water" to sustain a nuclear reaction. During nuclear reaction, neutrons from the uranium fuel change some of the reactor's heavy water into tritium. This radioactive tritium remains in the heavy water and builds up over time until it is physically removed at a tritium removal facility. In result of thermonuclear weapon test at 1959–1962 years the tritium ($1{,}82 \cdot 10^{20}$ Бк) was emitted in the atmosphere. The tritium concentration in the rain water has increased in 10-100 time after hydrogen bomb explosion [1]. Tritium is the most commonly encountered and important beta-emitting radionuclide. It is an internal radiation hazard when inhaled, or ingested via food or water, or absorbed through the skin. Radioactive (tritiated) water is generally indistinguishable from normal water and is transported throughout the environment in the same manner as water.

Radioactive decay of tritium produces what is known as ionizing radiation. According to the National Institutes of Health, Hazardous Substances Data Bank, Ionizing radiation has carcinogenic effects in many tissues. A dose response relationship exists between exposure to ionizing radiation and the risk for the subsequent development of cancer. This means the more radiation a person is exposed to, the greater their chances of developing cancer [5]. Ionizing radiation is genotoxic and causes breaks in the structure of DNA, resulting in mutations or chromosomal structural aberrations. Double DNA strand breaks that result in mutagenic and carcinogenic effects have been reported. Incorrectly rejoined DNA after broken DNA rejoins leads to DNA deletions and rearrangements. Large scale changes in DNA structure appear to be typical of most radiationinduced mutations. Therefore the distribution of tritium in the environment should be controlled.

M.A.Kulkova is with the Herzen State Pedagogical University, St.Petersburg, Russian Federation (corresponding author to provide phone: +7(812)3144796; fax: +7(812)3144784; e-mail: kulkova@ mail.ru).

A.B.Davidochkina is with the Herzen State Pedagogical University, St.Petersburg, Russian Federation (e-mail: skyrocketing@mail.ru).

II. Radioactive Pollution Of Gulf Of Finnland Basin

At present the specialist-ecologists devote the much attention to ecology of Baltic Sea basin. The countries located around Baltic Sea are under disaster of radionuclide pollution. The Baltic Sea is shallow and has isolation from Atlantic Ocean. It is the reason of low capability for purification and the time of whole water exchanging come to 27 years [8]. The industrial waste from nine countries which has on their coasts the nuclear-power reactors falls into Baltic Sea. The first disaster of the radioactive waste on the background of global radioactive pollution has appeared after damage of Baltic nuclear-power reactors. In this period the radioactive nuclides from Nuclear Power plants of Western Europe (Sellafield, La Hague) came in Baltic Sea through Danish Strait. On the data of HELCOM [4,7] there are twelve Swedish, four Finnish and nineteen Germany power(-generating) units in force in the Baltic Sea zone. In the Gulf of Finnland or Finnish Bay the Leningradskaya (Sosnovii Bor) Nuclear Power Plant is situated. The Gulf of Finland (Fig.1) is the easternmost arm of the Baltic Sea. The eastern parts of the Gulf of Finland belong to Russia, and some of Russia's most important harbors are located farthest in, near Saint Petersburg. Some of the environmental problems affecting the Baltic Sea are at their most pronounced in the shallow gulf. At the area of Nuclear Power plants the depositories of radioactive waste is located. The atomic submarines and ships are repairing on the coastal parts. Today the potentially dangerous sources of man-caused radioactive nuclides in the environment of Baltic Sea amass at Leningradskaya Nuclear Power Plant, at Kola Nuclear Power Plant, at Ignalinskaya Nuclear Power Plant (Lithuania).

The numerous radiation-dangerous objects are concentrated within the St.Petersburg city and around one. This is the objects of medicine, shipbuilding, scientific investigations and others. The Chernobil accident had impact on the ecology of Baltic Sea basin too. The investigations of heavy radioactive nuclides in the environment of Baltic Sea

region have a regular character. On the contrary at present the data about distribution of such long-living isotopes as tritium in the Baltic Sea ecosystem is almost absent. So this radioactive isotope makes the most contribution of radiation dose during Power Plant operations. The systematic control of tritium concentration in the water of Baltic Sea was realized at 80-th, begin 90-th years. The monitoring at 1972 year revealed that tritium concentration in the surface water of Ocean was considerably lower than in the surface water of Baltic Sea. The tritium concentration in the surface water of Baltic Sea at 1972 year was 47-70 TU (5,55 – 8,26 Bq/l), in the Finnish Bay - 62 TU (7,32 Bq/l). These results illustrate the pollution of Baltic Sea water is much greater than the Ocean water because in the Baltic Sea there are not active mixing of radioactive releases from atmosphere. It is important to control the level of tritium concentration in the water of Baltic Sea basin.

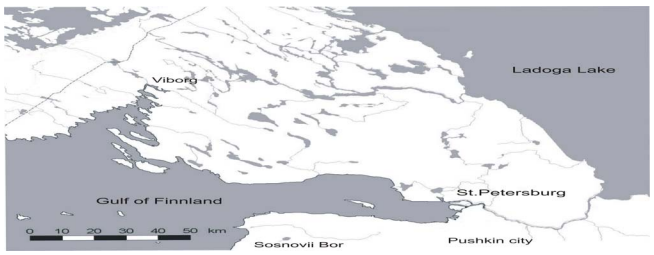

Fig.1 The map of Gulf of Finnland basin

III. Method Of Investigation

A. Sampling of water and vegetation samples

The sampling of water and snow was carried out from August 2008 year to June 2010 year. 70 samples (39 water samples, 11 snow samples and 20 vegetation samples) were collected on the territory of Gulf of Finnland. One sample was collected from Finnish Bay of Finland. Some samples were collected from Ladoga Lake.

B. Treatment and measuring of tritium samples

Determination of tritium at ultra-low concentrations is important in several applications. It is important, for example, for study of uptake, sorption of tritiated compounds in the environment. The low tritium concentration in the environmental water (lower than 1.0Bq/kg-H_2O) is within the detective limit by the low background liquid scintillation counter. As for the samples close to or lower than the detective limit, the samples have to be treated. The one of main parameters that should be tested during measuring is quenching. The quenching in one sample depends on the scintillation cocktail, the vial, and the sample treatment and amount. In practical work, there is only a need for knowing the amount of quenching, i.e. the counting efficiency, to enable correction of the measured count rates. To obtain the small variations in the quenching of different samples should be used the same amount of samples, the same scintillation cocktail etc. In the Quantulus counting the main procedure for establishing the amount of quenching in a sample is the spectrum quench parameter, SQP(E), based on external radiation. This means that an external gamma source irradiates the sample generating Compton scattering in the liquid solution. The shape of an unquenched spectrum is known by the detector and the SQP(E) is simply the ratio of the mass center of the measured spectrum to the unquenched one. To obtain the lowest level of detection (and highest figure of merit) the selection of cocktail and the mixing ratio is important [2,3,9].

Aqueous samples were prepared before measurements [2]. 100-500 ml of water was distilled to dryness. This removes dissolved salts that may cause quenching and also dissolved radon and progenies are removed. By distilling to dryness one avoids problems with fractionation of HTO and D2O over H2O. Aliquots of the distillate were taken out and added to vials. The vials manufactured from polyethylene (PE) were used. Optiphase Hisafe 3 cocktail was added. The ratios of water to cocktail 8:12 have been used [6]. The same amount of water from Gatchina underground sources was used for determination of the background count rate.

The system of sample preparation Sample Oxidizer 307 has been applied for solubilization of vegetation debris for liquid scintillation analysis [10]. The Liquid Scintillation Spectrometer Quantulus was used for the counting. It has an active guard in addition to a lead shield. The guard isoperated in coincidence with the true event signal from the sample's photomultiplier tubes (PMTS). The samples were counted in at least two cycles, usually for 120 min each time. By this means it is possible to see contributions from chemical luminescence in the tritium spectrum.

IV. Results

The results measurements of tritium in the water of the Finnish Bay basin are depicture on the Figure 2. This is average tritium concentration (TU) for different aqueous objects of Baltic Sea basin (rivers, snow, lakes, underground sources, St.Petersburg waterway, waste water and water of Baltic Sea).

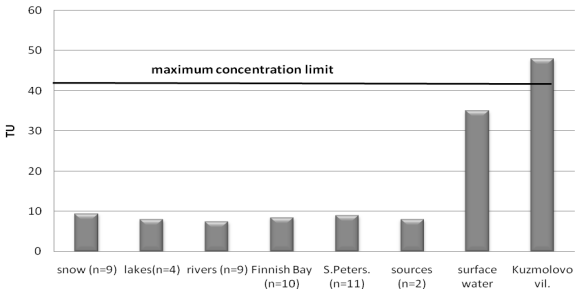

Fig.2 Distribution of tritium in different water objects of Finnish Bay basin

The most high of tritium concentration 48,8TU (5,77 Bq/l) was registered in the waste water of Kuzmolovskii village (Fig 3). This is little higher than the background concentration in the waters but it is within the range of maximum concentration limiting. This concentration is higher than tritium concentration in other groups of objects on average in 7 time and in 1,3 time it is higher in comparison to surface water from Oserki district of St.Petersburg city. Probably, the site of radioactive storage located near Kuzmolovskii village is the reason of high level of water radioactivity. In Oserki district of St.Petersburg city, near

Viborg highway (Fig 4) the high tritium concentration 35,6 TU (4,2 Bq/l) were marked. This can be connected with transportation of radioactive waste via city.

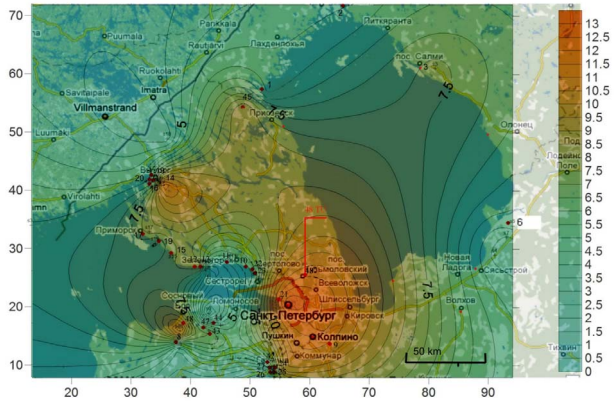

Fig.3 The map of total tritium distribution in the water on the territory of Finnish Bay basin

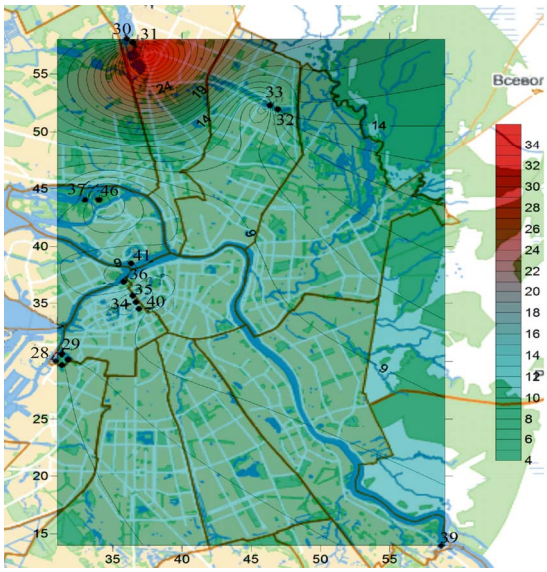

Fig.4 The map of total tritium distribution in the water on the territory of St.Petersburg city

An average tritium concentration in snow is 8,4 TU (1 Bq/l). This is more than in other water objects in 1,2 time. We supposed that it is atmospheric tritium, which falls out as snow. The tritium concentrations between water of rivers, lakes and sources of Baltic Sea basin have little differences and an average it is 5-7 TU (0,9 - 0,6 Bq/l). This is the same concentration as a water natural tritium background. The tritium concentrations from water of Finnish Bay and waterway of St.Petersburg have similar meanings, 7,1 TU (0,85 Bq/l) and 7,8 TU (0,92 Bq/l) respectively. It is much lower in comparison at 1972 year (62 TU (7,32 Бк/л)). The water sampled near Sosnovii Bor Power Plant has little higher tritium content, but it is not higher normal level. The water samples from lakes (Ladoga, Uzlovoe) and underground sources have similar tritium concentrations, 6,2 TU (0,73 Bq/l) and 6,7 TU (0,80 Bq/l) respectively. The concentration tritium from radon underground sources is low. The most low tritium concentration was registered in the rivers of district 5,1 TU (0,60 Bq/l).

The tritium concentrations in vegetation covers of Central district of St.Petersburg city and Pushkin city which are Southern suburb of St.Petersburg are depictured on the Figure 5.

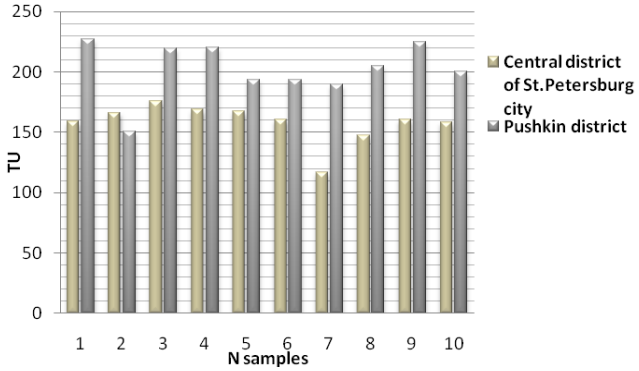

Fig. 5 The tritium activity in the vegetation covers

The tritium content in vegetation covers is higher in comparing to the water. In vegetation of Pushkin city the tritium activity is the most high, about 230 TU (27,15 Bq/l). The possible cause is the localisation of this district nearer to Nuclear Power Plant than the Central district. The data obtained accurately testify that tritium concentration in biological objects (organic connections) is higher than in the water in 5,5 times. In biological compounds the tritium has more strong organic connections than in the water. The vegetation receives the tritium both from air and water and soil.

The results obtained give the possibility to observe the tritium distribution in the different water objects and vegetation covers in the Environment of Gulf of Finnish of Baltic Sea and determine the tritium flux on the first stage of pollution. This is important for ecology of region. At present the level of tritium concentration in the water and vegetation of Finnish Bay Basin is in normal range and it has not risk for people health, but this level need be under control.

ACKNOWLEDGMENT

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