Estimation of Tritium Content in Atmosphere Using Lichen-indication Technique

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Abstract. Lichens are used for qualitative assessment of atmosphere pollution by heavy metals, radionuclide and gaseous pollutants. There are presented in the paper the results of the study to use lichens as quantitative tritium bio-indicator. The investigations were carried out in natural environment. Two experimental sites were used: one in vicinity of an emission source and another - at a distance. Air and lichen were sampled monthly. Both tritium in tissue free water (TFWT) and organically bound tritium (OBT) were measured in lichen. TFWT was extracted by thermal vacuum desorption. Vanadium oxide was used to oxidize dried lichen samples for OBT measurements. Air sampling was performed with aspiration technique. Liquid scintillation was used to measure tritium activity in water samples. There are presented in the paper the measurement results of both TFWT and OBT in lichens as well as HTO activity on atmospheric moisture for three years of observations. Based on experimental statistics there were determined regression relationships between the tritium activity in TFWT and organic matter in lichens and tritium activity in the atmospheric moisture.

Keywords: Atmosphere, Bio-monitor, Contamination, Correlations, Lichens, Tritium

1 Introduction

Application of epiphyte lichens for bio-monitoring is based on the fact that lichens absorbs all elements from the atmosphere. [1]. The surface of the lichen is a fungus micella and alga-cells [2]. In contrast to higher plants, this surface does not have a developed cuticle and stomatal apparatus, which act as a shielding and regulator barrier, able to control the process of gas and water exchange with the environment. Thus, lichen thallus is open 24 hour per day during dozens of years for elements of the environment. Moreover, the size of the open sorption surface of the lichen is 20-100 times greater than that of the superior plants and herbals, which is stipulated by the lichen texture [3]. A great number of research works is devoted to lichens use for qualitative assessment of atmospheric contamination by heavy metals, radionuclides, sulfur oxides, nitrogen etc. [4], [5].

The problem of using lichens to study the atmosphere pollution by tritium is not sufficiently studied. There is some research on studying the lichens capability to oxidize atmospheric molecular tritium [6]. It is shown that the rate of this process within a lichen substance is 100-1000 times greater than that of the pine needles and can be from 3 to 10 percent in hour. The authors have made a conclusion about perspectives of using lichens as monitors of the environment, specifically in order to detect the ways of movement of elementary tritium “cloud”. Earlier we indicated that the tritium content in lichen samples (both TFWT and OBT) in vicinity of emission source is higher than that of tritium content in lichens at distant sampling sites [7]. At that, TFWT content was in equilibrium with HTO content in atmospheric moisture, whereas the OBT content in lichens is considerably higher. It was supposed that TFWT content in lichens is related to the content of tritium in atmosphere at the minute of sampling, while OBT content was determined by tritium content in atmosphere over a long period. At the other research there were establish that compared to the lichens taken from the background location, the lichens of contaminated areas show very elevated concentrations of OBT, some of them is near 10 KBq·l^{-1} [8]. In addition, the lichens are unique in their tritium fixation capacity; the integration of HTO is occurring all around the year and is independent of ground water [9].

In order to use lichens for qualitative assessment of atmospheric contamination, it is essential to determine the correlation dependence between the tritium content in lichens and its content in atmosphere. The present work is devoted to the study of possibility to use the lichens for qualitative assessment of atmospheric contamination by tritium.

2 Experiment setup

Investigations were carried out in the central region of Russia. There is a constantly operating source of tritium emission in the area of investigation. The period of this source operation is more that 40 years. Two sites were chosen, one being near the source of tritium release in to atmosphere and another being at a distance of about 30 km, opposite to predominant wind direction. The sites were conventionally called “clean” and “contaminated”. The area of sites was about (100×100) m. The prevalent type of vegetation at the sites is a mixed forest. The main kinds of trees are pine tree, birch tree, aspen tree, linden tree and fir tree. Each site was equipped with air sampling stations for tritium content. Lichen specie Hypogimnia physodes (L) Nyl. was used as a basic bio-indicator. The lichen samples ware taken once a month. Lichen sampling was made from the trees trunks at the height of about 1.5 m from the earth along the entire length of the tree circuit. The samples were taken from all the territory of the experimental site uniformly. Both TFWT and OBT were measured in lichens. To extract TFWT, the method of thermal vacuum desorption was used [7]. The desorption temperature was +100˚C. Isolation of OBT from the dry residue of lichen sample was made at the same facility, using pyrolytic decomposition in the presence of vanadium oxide (V) at +500˚C for organic matter oxidation up to structural water and carbon oxide.

Air was sampled by pumping the air through the water vapor absorber [10]. A synthetic zeolite of the type NaA was used as a sorbent. Oxidation of HT up to HTO occurred on the platinum catalyzer, applied to aluminum oxide. The tritium content in air samples was measured once a month. Isolation of water from zeolite was performed by thermal vacuum desorption at +300˚C. Tritium content in the water extracted from the lichen and zeolite was measured with a liquid scintillation radiometer. The experiments were conducted during 30 months.

3 Results and discussion

Received during all period of observation the average values as well as the tritium content in lichens and air, are given in Table 1, Fig.1, 2.

Table 1. Average values of tritium content in air and in lichens.

The determined parameter	Tritium content, Bq l^{-1}	
	“clean” site	“contaminated” site
HTO in the air	67.0	947
TFWT in lichens	125	599
OBT in lichens	233	1720

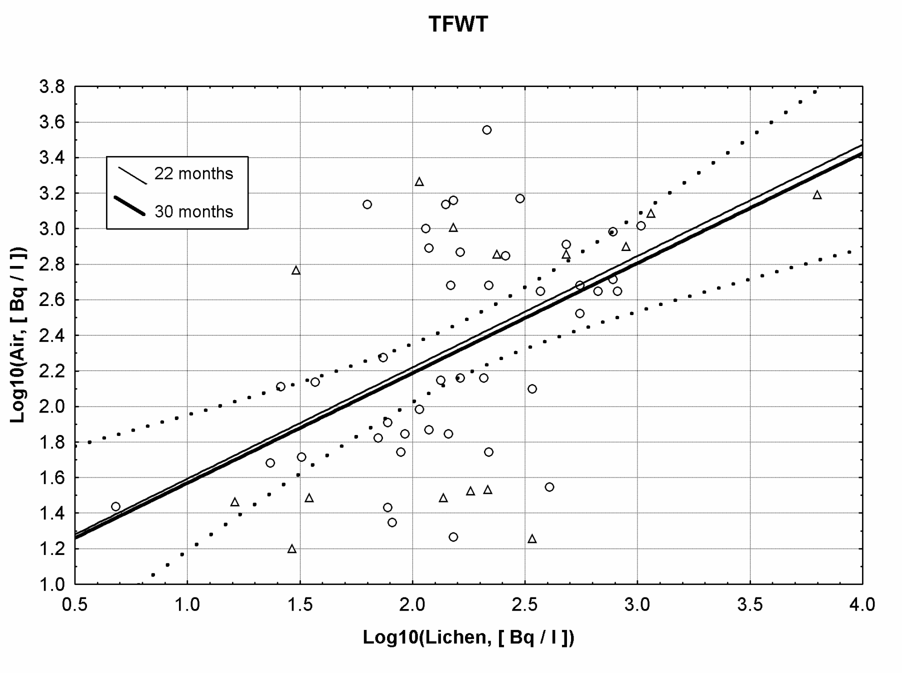

Figure 1. Tritium content in the air moisture and TFWT lichen and regression relationships.

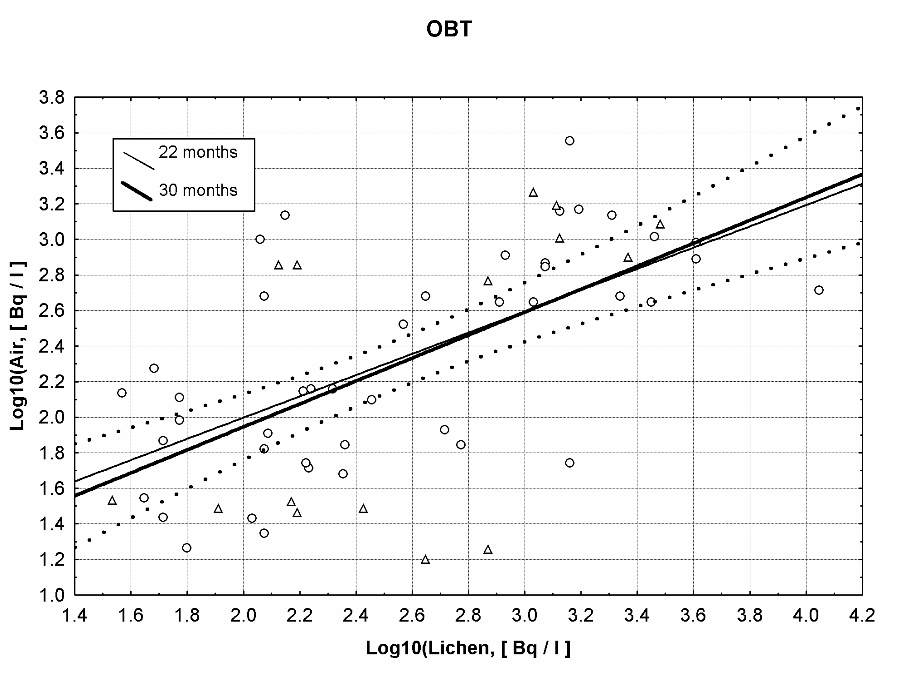

Figure 2. Tritium content in the air moisture and OBT lichen and regression relationships.

On the diagrams, the logarithm of HTO content in the air moisture is put along the X-axis, and the logarithm of TFWT and OBT content in the lichen – on the Y-axis.

The results showed that the content of HTO in atmospheric moisture and in lichens does not exceed the legitimate value of tritium content in water, which is $14.8 \cdot 10^3$ Bq l^{-1}.

The primary processing of measurement data has shown that samples, in most cases, do not meet the normality criteria, that hampers the use of traditional t-criterion. Thus, during the analysis of differences of tritium content in lichens and in atmosphere for "clean" and "contaminated" sites the well-known non-parametric tests of difference between the independent groups were used. These are criterion of Wald-Wolfoviz, Mann –Whitney U-criterion and Kolmogorov-Smirnov two-sample test [11]. All criteria indicated the presence of statistically significant difference in the content of tritium in air and lichens, sampled at "clean" and "contaminated" sites. Basing on the experimental data, received during the overall period of observation 30 months and during 22 months there were constructed the dependences between TFWT and OBT content in lichens and tritium content in atmospheric moisture there were constructed the dependences between TFWT and OBT content in lichens and tritium content in atmospheric moisture, Fig. 1, 2. The confidence interval (P=0.95) for 30 months regression line was determined (is marked by dot line). The dependences, constructed for 30 months are:

for TFWT lichen:

$$C_{AIR} \approx 10^{-3.08} \times C_{TFWT}{}^{0.62} \approx 8.26 \times 10^{-4} \times C_{TFWT}{}^{0.62}, \quad (1)$$

for OBT lichen:

$$C_{AIR} \approx 10^{-3.09} \times C_{OBT}{}^{0.65} \approx 8.09 \times 10^{-4} \times C_{OBT}{}^{0.65}, \quad (2)$$

where C_{AIR} is tritium content in atmospherics moisture (Bq l^{-1}), C_{TFWT} and C_{OBT} are content of TFWT and OBT in lichens (Bq l^{-1}).

There has been studied the possibility of lichen-indication method using for monitoring of atmospheric contamination by tritium using "contaminated" site. For that the 22 months regression dependences were used, Fig. 1, 2. During the check of this method there have been used the data, received in last 8 months. For the values of TWFT and OBT content in lichens measured in last 8 months, the HTO content in atmosphere moisture was determined, using the regressive dependences, Eq.(1), (2). The received data were compared with those, measured at air sampling station. The comparison of the values is given in Table 2.

Table 2. Comparison of the tritium determination results in the atmosphere moisture by means of lichen-indication and by means of air sampling stations in "contaminated" site

Sampling station, Bq·l^{-1}	TFWT Lichen-indicacion method, Bq·l^{-1}	Difference between the sampling station and TFWT lichen-indication method		OBT Lichen-indication method, Bq·l^{-1}	Difference between the sampling station and OBT lichen-indication method	
		In absolute units, Bq l^{-1}	In relative units [1)] (> or < *n* times) [1)]		In absolute units, Bq l^{-1}	In relative units (> or < *n* times)
722	444	*278*	*> 1.63*	130	*592*	*> 5.57*
796	651	*144*	*> 1.22*	651	*144*	*> 1.22*
1550	2220	*-670*	*< 1.43*	459	*1091*	*> 3.38*
1850	173	*1677*	*> 10.7*	411	*1439*	*> 4.50*
1020	215	*805*	*> 4.74*	466	*554*	*> 2.19*
722	285	*437*	*> 2.54*	118	*603*	*> 6.11*
1220	766	*454*	*> 1.59*	766	*454*	*> 1.59*
588	78.4	*510*	*> 7.50*	329	*259*	*> 1.79*

[1] The ">(<) n" means that the value measured with sampling station is n times greater (lesser) than that measured by lichen-indication method.

The results obtained by means of two methods differ, for the most part, 1.2-7.5 times for TFWT and 1.2-6 times for OBT. The exceptions are those measurements, when the difference for TFWT - 10.7 times (are marked in Table 2).

It is interesting to note some tendencies.

- The 22 months regression line is within the confidence interval for the 30 months regression line.
- The lichen-identification method, both for TFWT (in most cases) and OBT gives the lower values of tritium content in the atmosphere moisture.

- For the “contaminated” site, the difference between the results of sampling station and lichen-identification method are smaller if using TFWT.

It is probable, that this is associated with biological processes in lichens, absorption and distribution of atmosphere moisture, containing tritium in lichens.

Upon the whole, in measuring tritium in the atmosphere the lichen-identification method gives good results. This method may be used for diagnostics of atmospheric pollution by tritium and reveal of contamination source.

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