



Using of Lichens to Monitor Tritium Contamination of Atmosphere

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Abstract. It is known that 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 during three years 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. It was shown that tritium activity in lichen samples in the vicinity of emission source is higher than that of tritium activity in lichen at distance. 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. (Brown & Beckett, 1985). It is shown that the lichens get nutrients for their growth and metabolism from wet and dry precipitations (Nash III & Egan, 1988; Nash III, 1996; Rodrigo et al., 1999), which penetrate into the lichens thallus mainly in the form of cations and anions from the solutions, existing in the environment (Kershaw, 1985; Nash III, 1996). The surface of the lichen is a fungus micella and alga-cells (Gollerbakh, 1977). 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 (Ramzayev, 1984). That is why the lichens absorb great quantities of solution, containing mineral compositions and return the absorbed water during drying up, accumulating the solutes in their thallus as quickly as they absorb the water. All this result in the fact that the lichens are able to accumulate the elements in the contaminated areas, and the quantity of which greatly exceeds their physiological need. The quantity of these elements, absorbed by the lichens is much greater than the in higher plants and herbals (Loppi et. al., 1997). 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. (Conti & Cecchetti, 2001). However, 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 (Ichimasa et al., 1989). 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 ~ 3-10%/h. 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 (Golubev et al., 2002) that the

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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. 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. 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.

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 than 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 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 were 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 (Belovodsky et al., 1985). 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 (Belovodskiy et al., 1985). 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 in 1999-2001 during 30 months.

3. Results and discussion

The measurement results of TFWT and OBT content in lichens and tritium content in atmospheric moisture for “clean” and “contaminated” sites are presented in Fig. 1-Fig.3. The average values, received during all period of observation are given in Table 1. The results of research work 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 $14.8 \cdot 10^4 \text{ Bq l}^{-1}$.

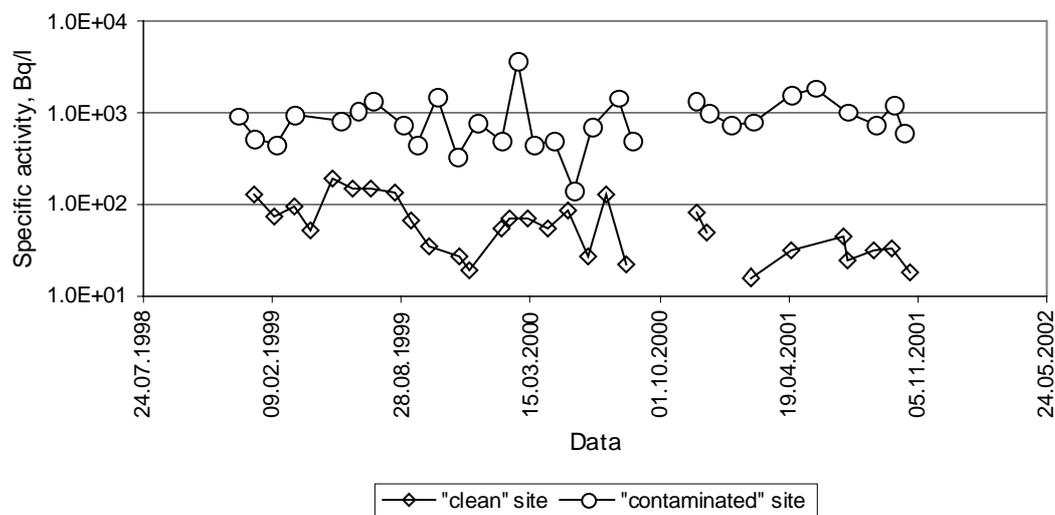


Fig. 1 Specific activity of tritium in the air moisture of “clean” and “contaminated” sites.

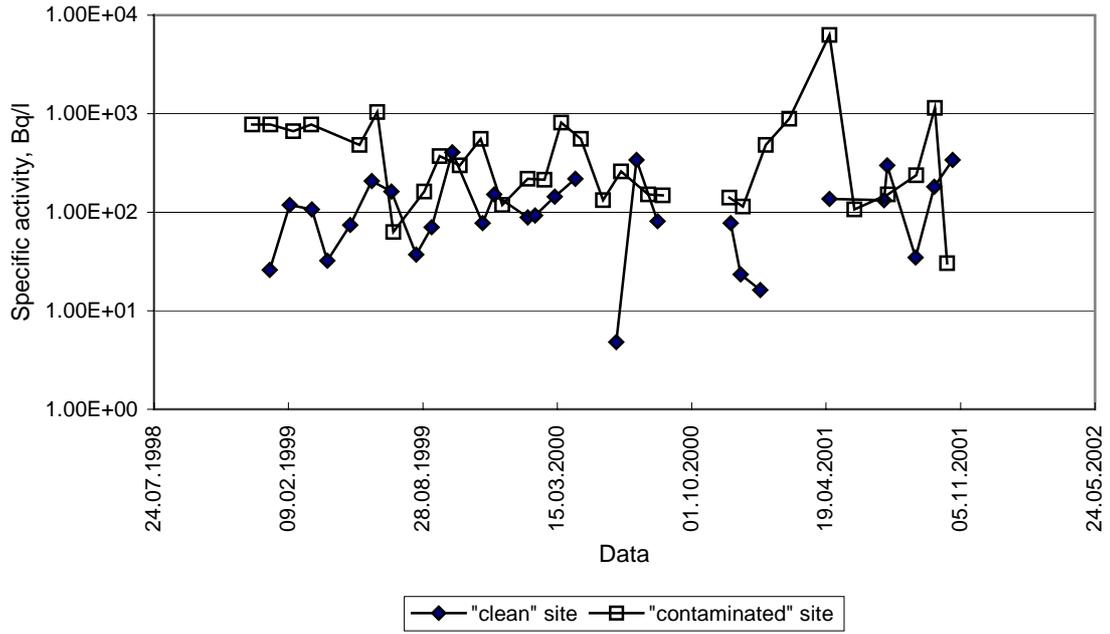


Fig. 2 Specific activity of TFWT of lichens from “clean” and “contaminated” sites.

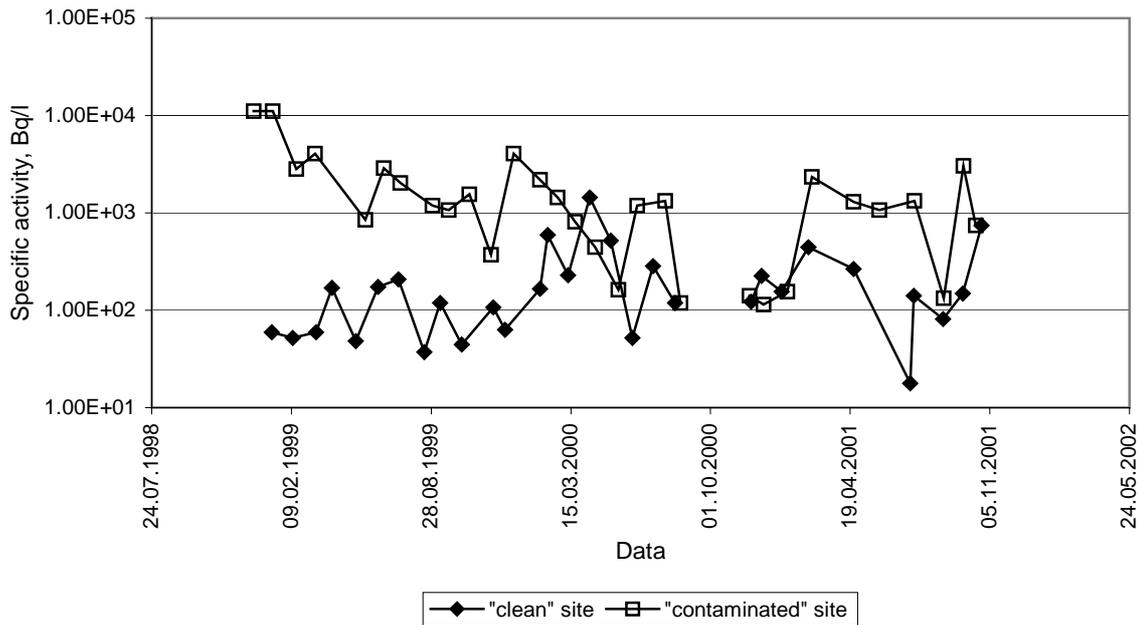


Fig. 3 Specific activity of OBW of lichens from “clean” and “contaminated” sites.

Table 1 Average values of tritium content in air and in lichens for “clean” and “contaminated” sites.

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

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 (StatSoft Inc., 2000). 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 in 1999-2001 and during 1999-2000 there were constructed the dependences between TFWT and OBT content in lichens and tritium content in atmospheric moisture, Fig. 4, and Fig. 5. 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 confidence interval (P=0.95) for 1999-2001 regression line is marked by dot line.

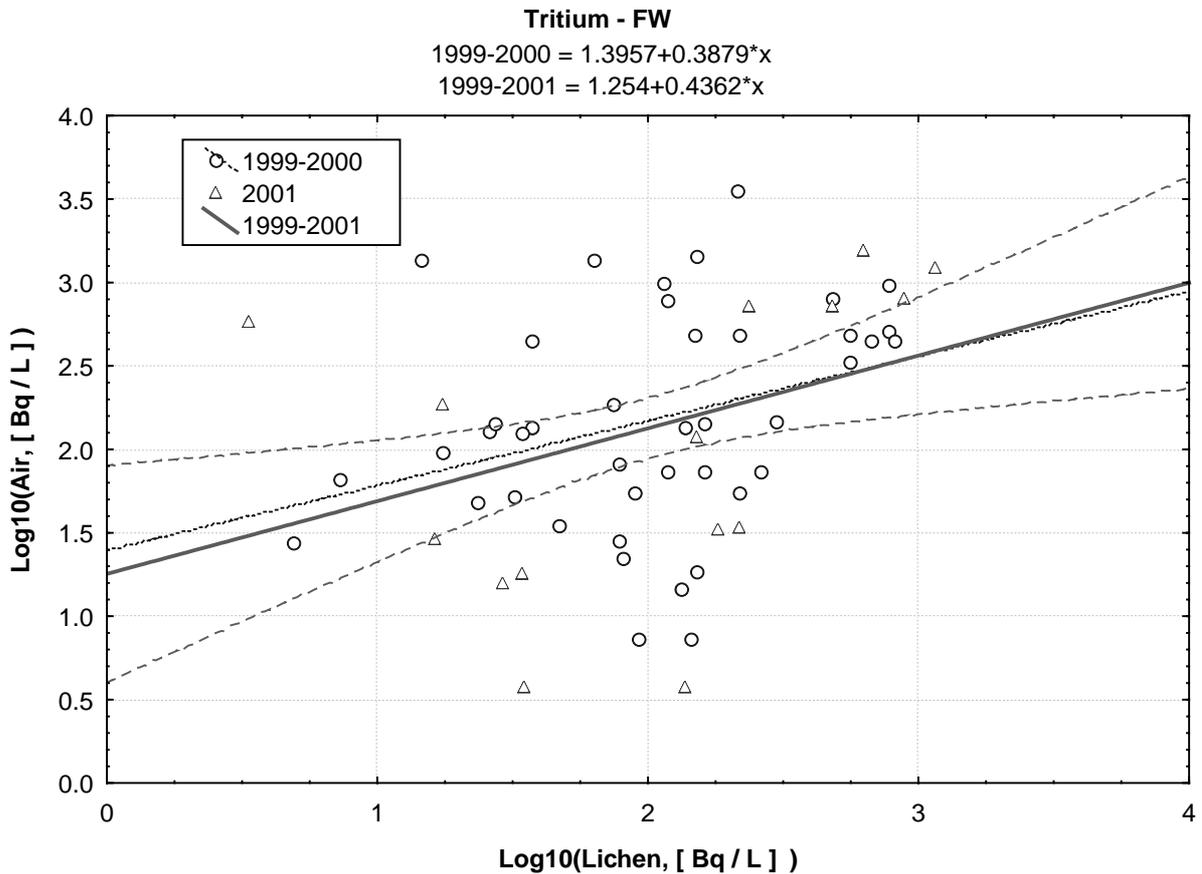


Fig. 4 Regression relationships between tritium content in the air moisture and TFWT lichen.

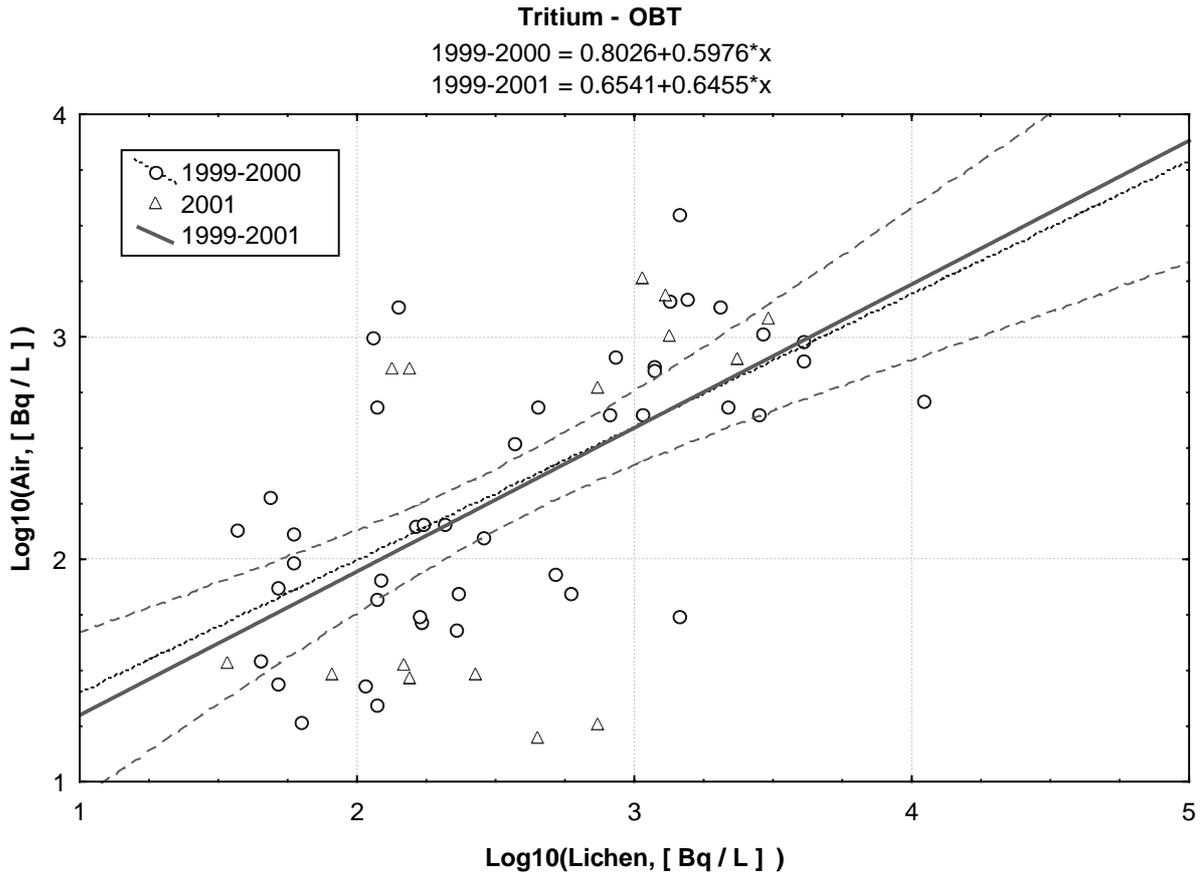


Fig. 5 Regression relationships between tritium content in the air moisture and OBT lichen.

The dependences, constructed for 1999-2001 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 atmospheric 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-identification method using for monitoring of atmospheric contamination by tritium. For that the regression dependences 1999-2000 were used, Fig. 4 and Fig. 5. During the check of this method there have been used the data, received in 2001. For the values of TFWT and OBT content in lichens measured in 2001, the HTO content in atmosphere 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 Tables 2, 3.

Table 2 Comparison of the results of tritium determination by means of lichen-identification and by means of air sampling stations (for TFWT)

Tritium content in the air, Bq l ⁻¹		Difference between the sampling station and lichen-identification method		
Sampling station	Lichen-identification method	In absolute units, Bq l ⁻¹	In relative units (> or < n times) ¹⁾	
<i>“Contaminated” site</i>	722	444	278	> 1.63
	796	651	144	> 1.22
	1550	2220	-670	< 1.43
	1850	173	1677	> 10.7
	1020	215	805	> 4.74
	722	285	437	> 2.54
	1220	766	454	> 1.59
	588	78.4	510	> 7.50
<i>“Clean” site</i>	15.9	76.6	-60.7	< 4.81
	30.7	202	-171	< 6.58
	44.4	198	-154	< 4.47
	24.1	330	-306	< 13.7
	30.7	85.5	-54.8	< 2.78
	33.7	241	-207	< 7.15
	18.1	357	-339	< 19.7

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

Table 3 Comparison of the results of tritium determination by means of lichen-identification and by means of sampling stations (for OBT)

Tritium content in the air, Bq l ⁻¹		Difference between the sampling station and lichen-identification method		
Sampling station	Lichen-identification method	In absolute units, Bq l ⁻¹	In relative units (> or < n times)	
<i>“Contaminated” site</i>	722	130	592	> 5.57
	796	651	144	> 1.22
	1550	459	1091	> 3.38
	1850	411	1439	> 4.50
	1020	466	554	> 2.19
	722	118	603	> 6.11
	1220	766	454	> 1.59
	588	329	259	> 1.79
<i>“Clean” site</i>	30.7	179	-148	< 5.82
	34.2	52.5	-18.3	< 1.54
	30.7	88.1	-57.4	< 2.87
	33.7	126	-92.1	< 3.74
	18.1	329	-311	< 18.1

The results obtained by means of two methods differ, for the most part, 1.5-8 times for TFWT and 1.5-6 times for OBT. The exceptions are those measurements, when the difference for TFWT -10.7-19.6 times, for OBT – 18.1 times (are marked in Tables 2, 3).

It is interesting to note some tendencies.

- The lichen-identification method, both for TFWT (in most cases) and OBT gives the lower values of tritium content in the air for “contaminated” site and the higher values for “clean” site.
- For the “contaminated” site, the difference between the results of sampling station and lichen-identification method are smaller if using TFWT.
- For “clean” site, the situation is reversed. The difference between the results of the sampling station and lichen-identification method are smaller if using OBT.

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

Upon the whole, in measuring tritium in the air 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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