The Quantitative Assessment of Uranium Atmosphere Pollution Using of Lichens

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Abstract. In bio-monitoring practice lichens are widely used for the assessment of radionuclide atmosphere pollution. In literature there were published dates on usage of lichens for the qualitative atmosphere control. There are presented in the paper the results of the study to apply lichens as a quantitative uranium bio-monitor of atmosphere contamination. The observations were carried out during 34 months in natural environment. Two experimental sites were used: one was in vicinity of an emission source of uranium and another - at a distance. Air and lichens were sampled monthly. Lichen species *Hypogimnia physodes* was used as a basic bio-indicator. The content of uranium in lichen and air samples was measured with mass-spectrometry after chemical extraction of uranium. There are presented the results of uranium measurements both in atmosphere and in lichens obtained during 34 months, the regression relationships between the content of uranium in lichens and in the atmosphere and the results of the lichen-indication technique testing. There are estimated a natural scattering of uranium content in lichens within the experimental sites. The rations of maximum to minimum uranium content in lichens are 4 and 1.5 for the experimental sites.

Keywords: Atmosphere, Bio-monitor, Contamination, Correlations, Lichens, Uranium.

1 Introduction

Lichens were first used as bio-monitors in 1866 during the description of Paris flora, when the contaminated air sensibility of these plants was noticed [1]. By the present time a great experience of using lichens as bio-monitors to study radionuclide atmospheric contamination has been gained [2]. The lichens were used for radionuclides fall-out assessment after the accident at Chernobylskaya nuclear power station [3]. In the outskirts of Saint Petersburg there has been detected ^{134}Cs in lichen *Parmelia sulcata* samples, gathered in summer 1991 [4]. The data agree with radioactive contamination of soil on this territory. In some regions of Norway the content of ^{134}Cs and ^{137}Cs in lichens is two times higher than that of vascular plants [3]. To assess the content of ^{129}I and ^{36}Cl in the regions near to Chernobyl, *Parmelia sulcata* lichen was used [5]. The comparison of the obtained data with the lay out of regional distribution of these radionuclides revealed the positive correlation with the concentrations accumulated in lichens. It was discovered that the radionuclides content decreases naturally in lichens (self-purification). The average biological period of self-purification for ^{137}Cs in *Xanthoria parietina* lichen is 58.6 months. If compared to higher plants, lichens are known by high ionizing radiation tolerance both to the effect of low permanent and critical forms [6]. It is noticed that the radiation exerts influence on the content and density of lichen associations [7]. The conducted tests revealed the correlations between the radionuclide concentration in lichens and in environment,

however they did not revealed any correlations between radionuclide concentrations in lichens and in atmosphere. Earlier we showed the possibility to apply epiphytic lichens as monitors of quantitative atmospheric contamination with anthropogenic airborne uranium [8]. There were compared the air pollution results measured by the lichen-indication method and by the air sampling station technique. The results obtained by two methods differ by no more than 6 times [8]. The natural scattering of uranium content in lichens within the experimental site can influence on the lichen-indication method results. The present work is devoted to the study of using lichens for qualitative assessment of atmospheric contamination by uranium as well as to estimate an influence of the natural scattering of uranium content in lichens within the experimental site on the measurement results.

2 Experiment setup

Investigations were carried out in the central region of Russia. There are constantly operating and instantaneous sources of uranium emission in the area of investigation. The period of these sources operation is more that 40 years. Two experimental sites were chosen, one being near the source of uranium release in to atmosphere and another being at a distance of about 30 km, opposite to predominant wind direction, which were conventionally called "clean" and "contaminated". The area of sites was about 100×100 m. The 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 monitoring stations for air aerosols sampling for uranium content. Lichen specie *Hypogimnia physodes (L) Nyl.* was used as a basic bio-indicator.

The lichens were sampled once a month. Lichen sampling was made from the trees trunks at the height of about 1.5 m from the ground along the entire length of the tree circuit. The samples were taken from all the territory of the experimental site uniformly. Further, the lichen samples were washed with twice-distilled and were mineralized by nitric acid at gradual heating up to $+(350\pm50)^0$C. Air sampling was made by continuous pumping of the air through the filters. The method of sampling was developed in compliance with IAEA recommendations [9].

The uranium content in air filters and in carbonized lichen samples was determined by means of mass-spectrometric isotopic dilution after uranium preliminary extraction. High-increased ^{233}U was used as a standard (isotope content – 99.9728 %at.). A fixed amount of standard was introduced into the samples, being analyzed. The extraction of uranium from the ash samples was done by ethylacetate from 1 M HNO_3, saturated with ammonium nitrate. The uranium isotopic ratios were measured by means of mass-spectrometer with surface ionization source. Analytical method is described in article [8].

The experiments were conducted during 34 months.

3 Results and discussion

The measurement results of U content in lichens and atmosphere for "clean" and "contaminated" sites are presented in Fig. 1, 2. The received during all period of observation average values are given in Table 1. The obtained results showed that the content of U in atmosphere does not exceed the values, stated by Canadian scientists as a limiting value for natural U: 0.48 microgram $^{-3}$ - for the health of a critical group of population and 0.075 microgram m^{-3} - as phyto-toxic from the point of view of ecology [10].

Table 1. Average values of U content in air and lichens for "clean" and "contaminated" sites [8]

Subject of investigation	U content	
	"Clean" site	"Contaminated" site
Air, microgram m^{-3}	0.0000113	0.000209
Lichens, milligram kg^{-1}	0.106	1.45

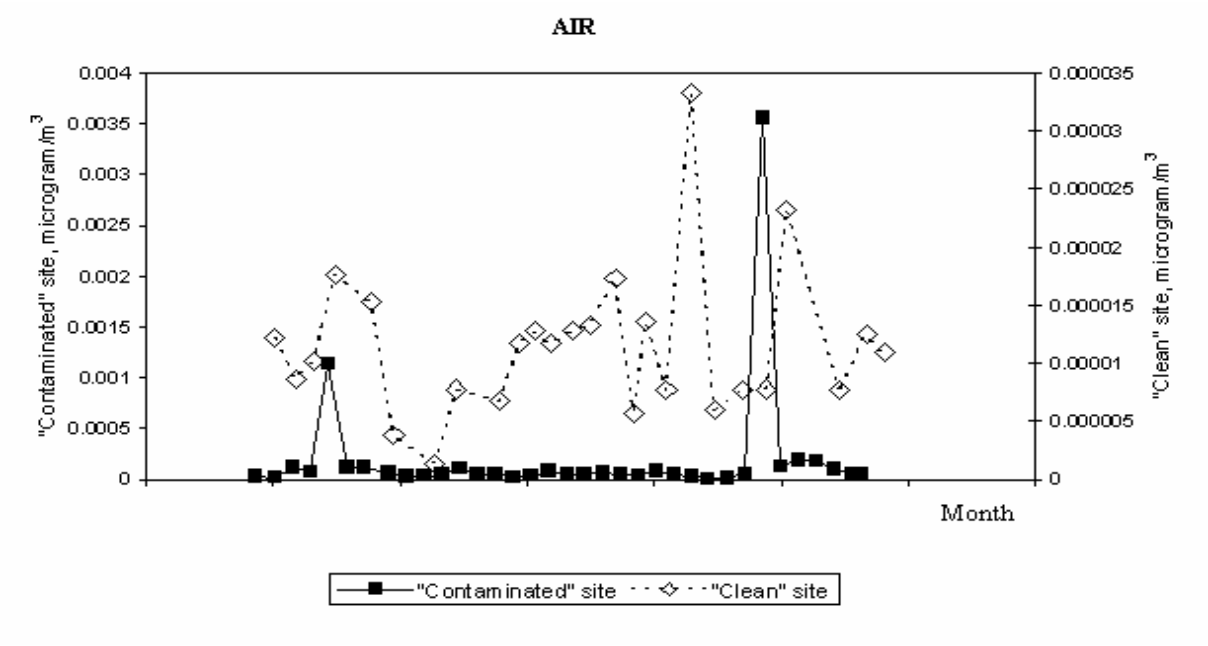

Figure 1. Uranium content in the air for “clean” and “contaminated” sites.

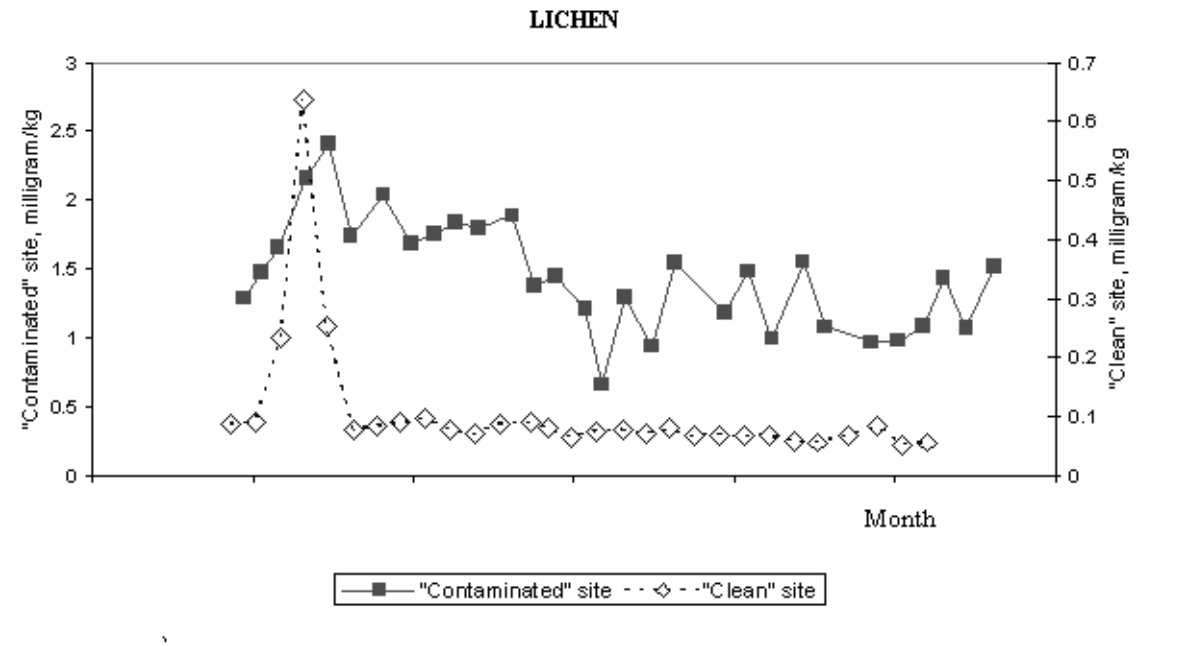

Figure 2. Uranium content in lichens for “clean” and “contaminated” sites.

Basing on the experimental data, received during the overall period of observation in there were determined the dependence between U content in lichens and U content in atmosphere [8].

The dependence is

$$C_{AIR} \approx \exp\left(1.1 \times C_{LICHEN} - 12\right),$$

where C_{AIR} is U content in the air (microgram·m^{-3}), C_{LICHEN} is U content in the lichen (milligram kg^{-1}).

Using the “contaminated” site there has been studied the possibility to monitor the uranium atmospheric contamination by lichen-identification method [8]. For that, for the values of uranium content in the lichen measured in “contaminated” site, the uranium content in the air was determined, using the regression relationship. The received data were compared with those, measured at the air sampling station, Table 2.

Table 2. Comparison of the results of uranium determination by means of lichen-identification and by means of the air sampling station

Uranium content in the air, microgram m^{-3}	
Sampling station	Lichen-identification method
1.44E-05	3.05E-05
5.41E-05	4.92E-05
9.55E-05	3.02E-05
1.27E-04	2.73E-05
1.82E-04	4.46E-05
1.84E-04	3.09E-05
3.56E-03	2.70E-05

The maximum difference is revealed for the point, marked in Table 2. In this point, the lichen-indication method shows the value of U content in the air, which is approximately 130 times less than that of the sampling station. Obviously, this is related to the biochemical properties of lichens as well as to physical and chemical properties of uranium particles In addition the U concentration in lichen is averaged over longer period time as compared the air aerosol samples as well as the lichen samples also contain “older” contamination. The U absorption by lichens may be affected by meteorological conditions, such as wind direction and rate, humidity, which determine the time for particles being in the atmosphere. Moreover the differences may be caused by the natural scattering of uranium content in lichens within the experimental site.

To estimate the influence of the natural scattering of uranium content in lichens within the experimental site on the lichen-indication method results there were sampled and analyzed lichen samples from “clean” and “contaminated” sites. For that each of the experimental sites were divided into 11 sectors on whish 11 lichen samples were picked. The obtained results and their statistical processing are presented in Table 3.

The variation of the uranium content in lichens for the “contaminated” site is more significant that one for the “clean” site. Probably this is caused by the physical and chemical properties of the uranium particles. At the “contaminated” site the uranium particles can have a large size as well as can exist in the form being difficult for lichens capture and metabolism. This can be a cause of the difference between the uranium measurement results obtained by the lichen-indication method and by the air sampling station method, Table 2.

Table 3. The determing of the natural scattering of uranium content in lichens within the “clean” and “contaminated” sites. Statistical processing of results

Sample	U content, $mg \cdot kg^{-1}$	
	”Contaminated” site	”Clean” site
1	3.066	0.0938
2	3.642	0.0982
3	4.168	0.0978
4	2.411	0.0787
5	7.152	0.0654
6	2.184	0.0657
7	5.115	0.0913
8	2.292	0.0797
9	2.681	0.0844
10	2.48	0.0779
11	1.765	0.094
Statistical processing		
Amount	11	11
Minimum	1.765	0.0654
Maximum	7.152	0.0982
Median	2.681	0.0844
Average	3.359	0.0842
Standart deviation	1.596	0.0118
Variation, %	47.5	14.1

In general, the method of lichen-identification gives positive results during detection of uranium in the atmosphere and may be used for diagnostics of atmospheric pollution by uranium and reveal of contamination source.

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