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AIR POLLUTION STUDIES IN CENTRAL RUSSIA (TVER AND YAROSLAVL REGIONS) USING THE MOSS BIOMONITORING TECHNIQUE AND NEUTRON ACTIVATION ANALYSIS

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INTRODUCTION

Toxic metals belong to the most serious air pollutants affecting our environment. Among the approaches used to identify these compounds and quantify their influence is the use of mosses as biomonitors of metal deposition, as illustrated in a recent review by Wolterbeek [1]. Since 1970 mosses have been regularly used in large-scale monitoring surveys providing valuable information on the relative spatial and temporal changes of trace metal deposition in the Nordic countries [2] and subsequently in several countries in Western and Eastern Europe [3]. The very first results obtained in Russia, from the northwest areas (Kaliningrad, Leningrad, Pskov Regions and Kola Peninsula), were reported in the 1995/1996 European moss survey along with data from 28 other countries [4]. In the most recent 2000/2001 moss survey, involving 30 European countries and coordinated by the UNECE ICP Vegetation Coordination Centre at the Center for Ecology and Hydrology (UK), results from the central part of Russia (Tver, Yaroslavl and Tula regions) were reported for the first time in the framework of this survey [5].

The aims of the present study were to determine levels of trace elements in mosses from Central Russia, to compare them to the European data, and to obtain information on the spatial distribution of heavy metal concentrations in mosses in the form of maps in order to estimate the impact from different sources of pollution.

MATERIALS AND METHODS

Study area

The neighboring Tver and Yaroslavl regions are located to the north of Moscow and belong to Central Russia, according to administrative division. The regions have similar geographical and climatic conditions, but for historical reasons the Yaroslavl region is more economically developed. There are a large number of towns and inhabited localities with various industrial activities in the Yaroslavl Region, whereas the Tver region is more characterised by agricultural activities. The investigated area is of great importance for Russia. Two fresh-water reservoirs forming the water-supply system for Moscow, the capital of Russia and nearby towns, are allocated within the territory. Among the potential pollution sources affecting the environment are a thermal power plant located in Konakovo on the shore of one of the fresh water reservoirs, a metallurgical plant located in the town of Cherepovets located at the northern end of the Rybinskoe reservoir, and various industrial plants in Yaroslavl, especially the oil refining plant (fig.1).

Figure 1: Tver and Yaroslavl regions: Sampling network.
Sites 1 – 36, sampling 2000; sites 37 – 103, sampling 2001;
sites104 – 139, sampling 2002.
1 - Thermal power plant, **2** - Steel plant, **3** - Oil refining plant.

Sampling

The moss species *Hylocomium splendens* and *Pleurozium schreberi* were sampled during three periods: During the summer-autumn 2000 moss samples were collected around the Moscow see. In summer 2001 sampling was continued in the eastern part of the Tver region. In 2002 sampling of mosses covered practically the whole Yaroslavl region. A single site in the vicinity of the largest metallurgical plant in Europe was included in that sampling as well. The network of sampling comprised a total of 140 sites, evenly distributed over the investigated territory (fig. 1). The sampling procedure was similar to that used in the previous European project "Atmospheric heavy metal deposition in Europe" [4].

Sample preparation

In the laboratory the samples were cleaned from extraneous plant materials and dried to constant weight at 30°–40° for 48 hours. The samples were not washed and not homogenised. Green and green-brown moss shoots corresponding to approximately three years of growth, were subjected to analysis. Previous experience from the use of NAA in moss biomonitoring surveys has shown that samples of 0.3 g are sufficiently homogeneous to be used without homogenisation [6].

Analysis

The concentrations of elements in the moss samples were determined by a combination of instrumental neutron activation analysis (NAA) and atomic absorption spectrometry (AAS). For NAA moss samples of about 0.3 g were heat-sealed in polyethylene foil bags or packed in aluminium cups for short and long irradiation, respectively. NAA was performed at the IBR-2 reactor in the Frank Laboratory of Neutron Physics. The analytical procedures and the characteristics of the pneumatic system employed are described in detail elsewhere [7].

Two different procedures of analysis were employed. The first was a short irradiation of 3–5 min to measure gamma-activities of short-lived isotopes (Al, Ca, Cl, Mg, Mn, and V). After a decay-period of 5 to 7 min the irradiated samples were measured twice, first for 3–5 min and then for 10–15 min. A long-irradiation of 4–5 days was used to measure long-lived radionuclides. After irradiation the samples were re-packed and measured twice, first after 4–5 days for 40–50 min to determine As, Br, K, La, Na, Mo, Sm, U, and W and then after 20 days for 2.5–3 hours to determine Ba, Ce, Co, Cr, Cs, Fe, Hf, Ni, Rb, Sb, Sc, Sr, Ta, Tb, Th, and Zn. The gamma spectra of the samples were measured with a Ge(Li) detector with a resolution of 2.5–3 keV for the ^{60}Co 1332 keV line or an HPGe detector with a resolution of 1.9 keV for the ^{60}Co 1332 keV line was used.

The processing of the data and determination of the element concentrations were performed using certified reference materials and flux comparators with the help of the software developed in FLNP, JINR [8].

To determine Cd, Cu and Pb, moss samples of 1.0–1.3 g were digested with 20 ml of concentrated nitric acid in a 100-ml Ehrlenmeyer flask on a hot plate at 80°–85° for 18–20 hours. After cooling to room temperature the samples were filtered and demineralized water was added to a total volume of 50 ml. Analysis for Cd, Cu and Pb was carried out by flame atomic absorption spectrometry on a Perkin-Elmer Analyst-600 spectrometer in the Norwegian University of Science and Technology, Trondheim, Norway. All calibration standards and blanks matched the nitric acid concentration of the samples. The AAS determinations were done only on samples collected in 2001.

Quality control

The QC of NAA results were ensured by carrying out a concurrent analysis of the reference materials (RM) Lichen 336 IAEA (International Atomic Energy Agency) and standard reference materials SRM-1575 (pine needles) from the US NIST (National Institute of Standards and Technology). NORD DK-1 (moss reference sample) prepared for doing the calibration in the laboratories participating in the corresponding 1990 Nordic survey were also used. For long irradiation the three reference materials were packed together with 10-12 samples in each transport container.

The RM showing least deviation between measured and certified values of elemental content was chosen. Results for Mg, Hf, and Ta, for which no certified reference values were available, were calculated using the Nordic moss reference sample DK-1 [8]. Mo and W were determined by the absolute method based on nuclear constants [9].

The quality of the AAS data was checked by analysis of moss reference samples [10] at regular intervals.

RESULTS AND DISCUSSION

Table 1 shows minimum, maximum and mean elemental concentrations and median values for 34 elements determined in moss samples from 139 sites in the Tver and Yaroslavl regions. The local background levels for elements were calculated as an average of values from the 15 sites showing the lowest contents of pollutant elements, to the north of town of Tver. The last column of Table 1 shows values for a single moss sample collected near the Chrepovets plant. As follows from the comparison, the concentrations of most elements are several times higher at this sampling site than the local background level. In particular this is evident for elements associated with emissions from ferrous metallurgy and steel industry (Fe, 36 times; Cr, 15 times; Ni and Mo, 13 times; V and As; 8 times). Concentration of such elements W, Pb and Cd in moss collected near Cherepovets plans is more than 5 times higher than the background level. For elements which are not specifically associated with the above industry, the values do not exceed the background level significantly (Zn, Al, Cu, Mn; 3.8, 3.6, 2.1, and 1.7 times respectively).

Factor analysis

Factor analysis is a multivariate statistical technique which has been successfully used in environmental pollution studies to simplify large and complex data sets in such a way that it may create one or more new variables (factors) which make it possible to explain the original data sets [11,12].

Table 1. Element concentrations in mosses (mg/kg) collected from Central Russia.

Element	Min	Max	Mean	Median	Local Background	Cherepovets
Na	62	3315	245	180	86	245
Mg	307	3360	1170	1070	540	1030
Al	123	7700	845	635	230	820
Cl	45	2270	225	160	77	135
K	3140	19610	7610	7260	3950	5300
Ca	1240	12340	3410	3060	1615	2500
Sc	0.027	1.27	0.14	0.10	0.04	0.37
V	0.34	29	3.2	2.6	0.73	6.4
Cr	<0.2	27	1.5	1.1	0.26	4.1
Mn	45	2200	400	315	95	160
Fe	68	3690	550	410	165	6120
Co	0.05	3.5	0.41	0.33	0.11	1.0
Ni	<0.25	22	2.0	1.5	0.28	3.6
Cu*	3.2	9.2	5.1	4.8	3.6	7.8
Zn	13	85	34	30	20	72.0
As	0.05	1.12	0.22	0.18	0.07	0.65
Br	0.5	6.9	1.6	1.4	0.7	2.20
Rb	4.9	106	25	23	8.9	16.6
Sr	3.2	74	13	10	4.7	11.0
Mo	<0.04	2.1	0.24	0.20	0.045	0.56
Cd*	<0.03	0.82	0.27	0.26	0.07	0.4
Sb	0.04	0.63	0.11	0.10	0.05	0.25
Cs	0.05	1.04	0.22	0.17	0.08	0.18
Ba	4	131	27	22	8	25
La	0.14	8.0	0.7	0.5	0.2	1.0
Ce	<0.25	14.7	1.5	1.0	0.25	1.2
Sm	0.014	0.95	0.11	0.078	0.031	0.150
Tb	<0.002	0.113	0.012	0.009	0.002	0.011
Hf	<0.03	2.3	0.18	0.09	0.03	0.15
Ta	<0.003	0.26	0.020	0.013	0.004	0.120
W	<0.025	1.22	0.18	0.12	0.03	0.2
Pb*	2.1	12.2	6.0	5.8	3.1	21.5
Th	0.017	1.7	0.14	0.10	0.035	0.10
U	0.008	0.45	0.054	0.040	0.014	0.06

* determined by AAS

The purposes of the factor analysis in our case were to define the number of common factors (independent pollution sources), to calculate the factor loadings, and to determine the geographical distribution of each factor (factor scores).

Factor analysis was applied to the data set of 32 elements in 139 samples. Cu and Pb were not included and the Cherepovets site was not considered. Factor loadings after VARIMAX rotation and explained variances are presented in Table 2. Significant values of loadings in each factor are marked in bold.

From the knowledge of the element composition of each factor, the significance of each element in the factor, and factor scores at each site, the sources of different elements in the mosses could be characterised.

Seven factors were recognised, explaining 75% of the total variance:

Factor 1 is a complex factor representing a soil component with high loadings for crustal elements. The geographical distribution of factor scores could be attributed to the emission of coal fly ash from local boiler plants during coal combustion.

Factor 2 comprises the elements **W**, Sr, Ba, Br, **Cd**, Ca **Zn,** Mg, and Fe. The highest factor scores are observed around Yaroslavl reflecting impact from various industries (fig 2).

Factor 3 is characterized by the highest loadings of Cl, K, Ca and includes Na, Mg, Zn and Sr as well. These are essential elements in mosses.

Factor 4 is represented by Mo, Co, Mn, Ca, and Fe and may be associated with manufacturing industry in Kashin and steel industry in Cherepovets. The geographical distribution of factor scores reflects the gradient of element concentrations along a N-S transect from Cherepovets (fig 2).

Factor 5 includes **V**, Sb, Al, and **Ni.** The pattern of factor scores depicts the location of the thermal power plant in Konakovo, which is based on the use of petroleum products. The pattern of factor scores also pints toYaroslavl, presumably due to the location of an oil-refinery plant there (fig. 2).

Factor 6 with the Rb and Cs is difficult to explain. Possibly it can be connected with the natural content of these elements in mosses.

Table 2. Varimax rotated factor matrix. Factor loadings and explained variance.

	Factors						
	1	**2**	**3**	**4**	**5**	**6**	**7**
% of Variance	40.1	9.9	6.6	5.9	5.2	4.0	3.3
Cumulative %	40.1	50.0	56.5	62.4	67.6	71.6	74.9
Na	0.82	0.07	0.43	0.05	0.04	-0.06	-0.04
Mg	0.43	0.43	0.35	0.05	0.17	-0.27	0.03
Al	**0.64**	-0.03	0.07	0.09	0.44	-0.15	0.01
Cl	0.14	0.19	**0.81**	-0.01	0.08	-0.01	0.01
K	0.18	0.01	**0.77**	0.11	-0.23	-0.03	-0.14
Ca	0.16	**0.52**	**0.57**	0.40	0.06	-0.20	-0.05
Sc	**0.92**	0.23	0.13	0.05	0.17	-0.05	0.04
V	0.18	-0.06	-0.02	0.00	**0.75**	0.06	0.16
Cr	0.18	-0.02	-0.05	0.05	-0.08	-0.06	**0.88**
Mn	0.04	0.06	-0.12	**0.66**	0.07	-0.06	-0.15
Fe	**0.69**	0.36	0.29	0.36	0.23	-0.04	0.07
Co	0.38	0.18	0.22	**0.77**	0.03	-0.06	0.01
Ni	0.06	0.06	-0.05	-0.04	0.31	-0.01	**0.84**
Zn	0.20	**0.51**	0.47	0.12	0.30	0.12	0.12
As	**0.75**	0.21	0.30	0.11	0.20	0.12	-0.02
Br	0.25	**0.56**	0.24	0.23	0.39	-0.02	0.10
Rb	-0.12	-0.19	0.10	-0.05	-0.07	**0.87**	-0.07
Sr	0.21	**0.64**	0.33	0.04	-0.10	-0.17	-0.03
Mo	0.08	-0.02	0.20	**0.88**	-0.08	-0.04	0.22
Cd	0.16	**0.55**	0.15	0.14	0.12	-0.03	-0.07
Sb	0.23	0.24	-0.06	0.00	**0.57**	-0.02	-0.02
Cs	0.06	-0.08	-0.16	-0.09	0.09	**0.89**	0.00
Ba	0.50	**0.61**	0.13	0.09	-0.03	-0.23	-0.10
La	**0.73**	0.05	-0.04	0.26	-0.01	0.00	0.03
Ce	**0.78**	0.00	-0.05	0.15	0.01	0.03	0.15
Sm	**0.92**	0.15	0.08	0.07	0.24	-0.03	0.07
Tb	**0.91**	0.11	0.07	0.03	0.16	-0.03	0.09
Hf	**0.93**	0.03	0.09	0.04	-0.04	0.01	-0.02
Ta	**0.87**	0.32	0.03	0.00	0.09	-0.08	0.08
W	0.03	**0.81**	-0.23	-0.14	-0.02	-0.06	0.11
Th	**0.94**	0.22	0.10	0.02	0.08	-0.05	0.03
U	**0.84**	0.27	0.18	0.04	0.19	0.00	0.07

Factor 7 contains Cr and Ni again. The highest factor score is located near Konakovo and may be connected with the production of building materials. Rather high factor scores are also observed around Yaroslavl (fig. 2).

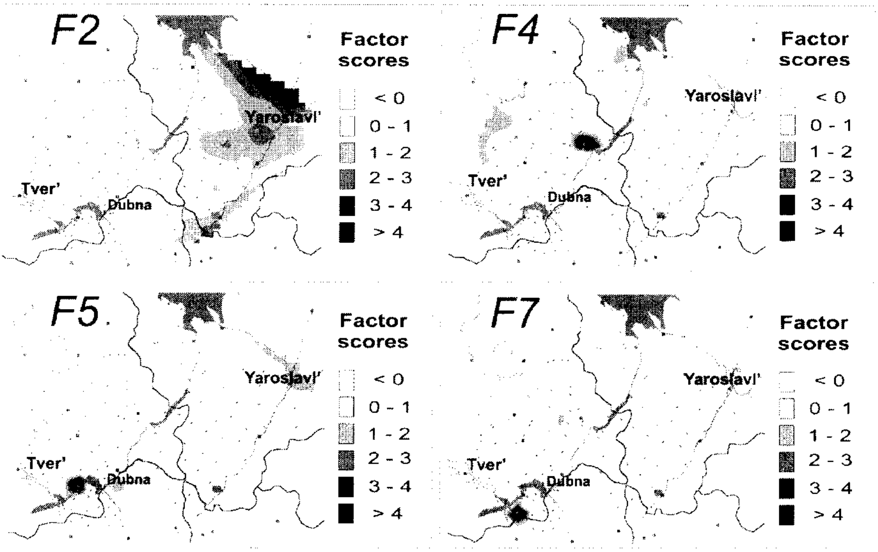

Figure 2. Geographical distribution of some factor scores
(GIS-INTEGRO was used to generate the maps)

Gradients of element contents in mosses

The three major anthropogenic sources of pollution were examined in detail. For this purpose moss data were chosen along a transect from Cherepovets approximately 300 km to the south, around Yaroslavl within a radius of 120 km to the southwest and around Konakovo within a radius of 50 km. Since there is no any prevailing wind direction, the temporal distribution may be similar in all directions from the pollution source. Element concentrations decrease rapidly with distance from the point sources, as evident from fig. 3, approaching the background level at 80 – 100 km distance. The observed curves can be represented by power functions:

$$y=ax^b \qquad (1)$$

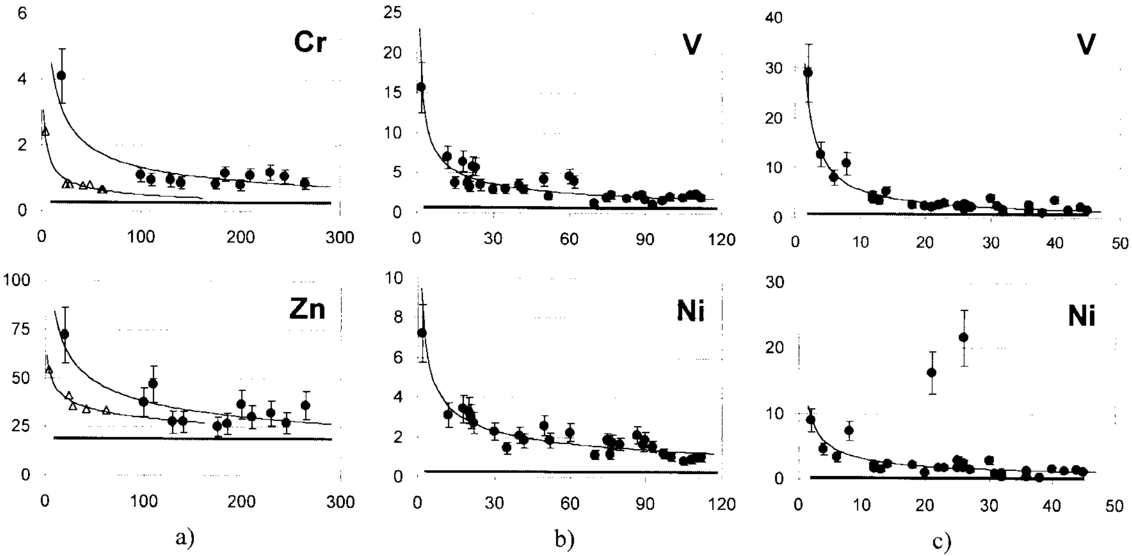

Figure 3. Element concentrations in mosses (mg/kg) versus distance (km) from the source:

a) along N-S transect from Cherepovets, b) around Yaroslavl, c) around Konakovo

Solid circles – present results, open triangles –1990, dashed curve is the background level.

The values of a, b and R^2 for each element are presented in Table 3. The value of b defines the slope of the curve. The elements specifically related to atmospheric emissions due to industrial activities have the highest values of b. Typical examples are Fe, Cr, As, Pb in the Cherepovets transect and V, Ni around Yaroslavl. The value of R^2 (coefficient of approximation) characterises the fitting of the curve to the experimental data. A high value of R^2 usually indicates a dominating impact from a single pollution source in the investigated area. The source of V in Konakovo (R^2=0.73) is a good example.

In the case of Ni there are two points deviating from the general trend expressed by the function. They correspond to Factor 7 as discussed above.

Table 3. The parameters of power curves.

					transect from Cherepovets 2000/2002					
	Fe	**As**	**Pb**	**V**	**Cr**	**Ni**	**Sb**	**Cu**	**Zn**	**Cd**
a	97700	4.3	136	27	15	12	0.88	18	184	0.8
b	-1.10	-0.60	-0.62	-0.51	-0.53	-0.46	-0.43	-0.28	-0.34	-0.26
R2	0.81	0.89	0.87	0.76	0.72	0.69	0.56	0.69	0.63	0.30
					transect from Cherepovets 1990					
			Pb *		**Cr ***	**Ni ***		**Cu ***	**Zn ***	**Cd ***
a			38		4	2		10	70	0.5
b			-0.46		-0.47	-0.16		-0.19	-0.19	-0.24
R2			0.87		0.94	0.85		0.63	0.95	0.68
					around Yaroslavl'					
	V	**Ni**	**Mo**	**Fe**	**Cr**	**W**	**As**	**Zn**	**Cd**	**Cu**
a	23	11	1.5	3250	6.3	1.1	0.7	90	0.6	9
b	-0.54	-0.47	-0.47	-0.43	-0.40	-0.38	-0.29	-0.24	-0.19	-0.17
R2	0.73	0.76	0.81	0.34	0.27	0.30	0.25	0.36	0.15	0.38
					around thermal power plant (Konakovo)					
	V	**Ni**	**As**	**Fe**						
a	44	15	0.3	625						
b	-0.90	-0.67	-0.31	-0.22						
R2	0.82	0.27	0.25	0.16						

Similar results are also shown from a previous investigation of moss samples collected in 1990 by the Institute for Biology of Inland Waters Russian Academy of Sciences along the same transect from Cherepovets, but 60 km farther to the south. The lower values for the main pollutant elements in that material are probably explained by a general industrial stagnation during that period. Nevertheless the elements decrease with distance from Cherepovets in a similar way.

Comparison of heavy metals atmospheric pollution in some areas of Russia

The participation of Russian teams in the 2000/2001 moss survey made it possible to carry out comparison of element contents in mosses from different areas of Russia. The median values for main elements pollutant in Tver, Yaroslavl as well as Tula [13] and St. Petersburg regions [5] are presented in fig. 4. As seen from the figure, both Tver and Yaroslavl are distinctly less polluted than Tula. Tver generally shows slightly lower medians than Yaroslavl.

The median concentrations of all heavy metals in regions of central Russia are similar to the median levels observed elsewhere in European countries participating in the 2000/2001 moss survey [5], as illustrated in fig. 5 in the case of V.

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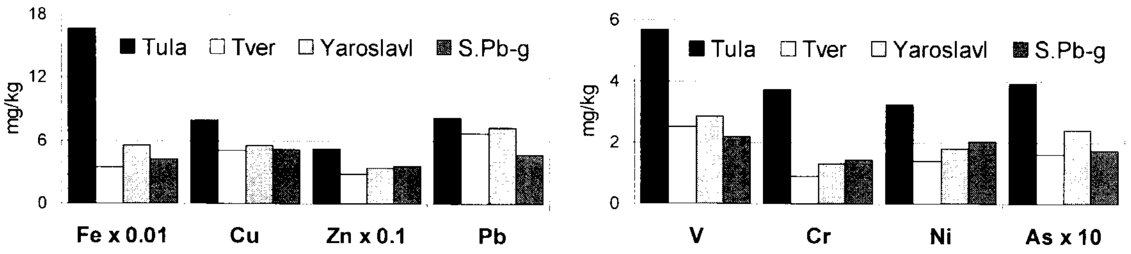

Figure 4. Comparison of median values of some metals in moss from different regions in Russia.

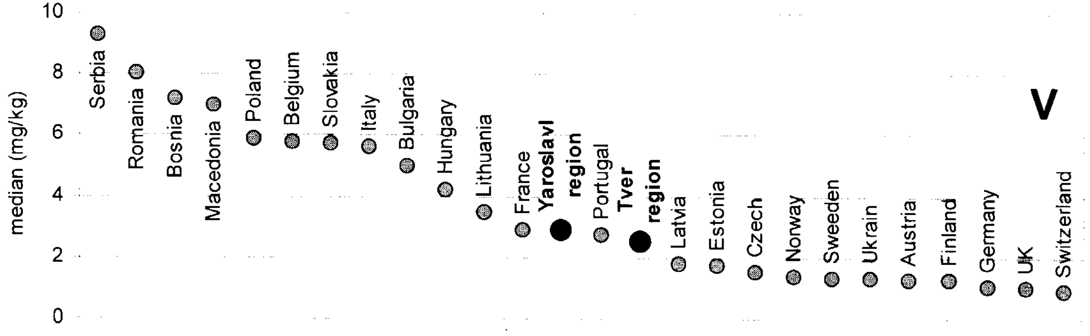

Figure 5. Median V concentrations in moss in different European countries.

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Ермакова Е. В. и др. E14-2003-196
Изучение воздушных загрязнений в Центральной России
(Тверская и Ярославская области) с помощью метода
мхов-биомониторов и нейтронного активационного анализа

Представлены данные 34 элементов, включая тяжелые металлы, галогены, редкоземельные элементы, а также уран и торий, обнаруженных в 140 образцах мха. Образцы мха были собраны в 2000–2002 гг. в центральной части России на территориях Тверской и Ярославской областей, а также на севере Московской области. Для определения возможных источников попадания элементов в мох применялся факторный анализ (VARIMAX-вращение). Семь результирующих факторов представляют собой почвенный, растительный и антропогенный компоненты во мхах. Антропогенные факторы интерпретируются как источник, связанный с металлургическим производством (Fe, Zn, Sb, Ta); сочетание разных типов производства (Mn, Co, Mo, Cr, Ni, W); нефтепереработка и сжигание нефтепродуктов на тепловой электростанции (V, Ni). Также представлено географическое распределение значений факторов. Приведены уравнения зависимости содержания элемента во мхах от расстояния от источника.

Работа выполнена в Лаборатории нейтронной физики им. И. М. Франка ОИЯИ.

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(Tver and Yaroslavl Regions) Using
the Moss Biomonitoring Technique and Neutron Activation Analysis

Data of 34 elements, including heavy metals, halogens, rare-earth elements, U, and Th in 140 moss samples, collected in central Russia (Tver and Yaroslavl regions and the northern part of Moscow Region) in 2000–2002, are presented. Factor analysis with VARIMAX rotation was applied to identify possible sources of the elements determined in the mosses. The seven resulting factors represent crust, vegetation and anthropogenic components in the moss. Some of the factors were interpreted as being associated with ferrous smelters (Fe, Zn, Sb, Ta); combination of non-ferrous smelters and other industries (Mn, Co, Mo, Cr, Ni, W); an oil-refining plant, and oil combustion at the thermal power plant (V, Ni). The geographical distribution patterns of the factor scores are also presented. The dependency equations of elemental content in mosses versus distance from the source are derived.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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