Monitoring of aerosol deposition flows in background areas of Tomsk Region

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Received January 29, 2007

The given study presents the results of spatiotemporal variability of aerosol deposition flows in the cold period for two background monitoring stations of IAO SB RAS, i.e., Akademgorodok and the reference one (the Kireevsk village, 60 km from Tomsk). Methods of stereoscopic binocular microscopy, X-ray structure, and instrumental neutron activation analysis were used when studying the mineral-matter and geochemical compositions of snow solid sediment samples. At the same time, the concentrations of submicron aerosol and soot in the surface air have been measured at the aerosol station of IAO SB RAS from November, 2005 to April, 2006. The differences in mineral-matter and geochemical compositions of snow solid sediment samples in two measurement points are revealed. An excess of aerosol flow values in Akademgorodok over those in the Kireevsk village is shown. A steady tendency to the decrease of the values of aerosol deposition flows in snow and concentrations of submicron aerosol and soot are established in the surface air in Akademgorodok when passing from winter to spring seasons.

Introduction

The petrochemical industry, enterprises of nuclear-fuel and fuel and energy cycles, such as State District Power Stations, Thermal Power Stations, etc., and many industrial enterprises in Tomsk Region produce emissions of considerable aerosol masses into the environment. Air mass transfer favors the propagation of aerosol emissions far outside the urban and industrial areas that can lead to pollution of vast territories and to the harmful ecological consequences. Therefore, to study the effect of large industrial centers on the environment, it is necessary to carry out regular investigations in the background areas.

The experience of operational in the Siberian Region has shown that snow sampling[1–3] is widely used to characterize air aerosol pollutions. The snow cover is an ideal deposited medium for accumulation and analysis of pollutants accumulated for the winter season. The particles of both natural and technogenic origins are fixed in the snow.

The given study presents the results of studying aerosol deposition flows by geochemical methods, as well as spatiotemporal variability of aerosol pollution distribution in two background areas of Tomsk Region.

Measurements and data processing

The snow was sampled at two monitoring stations of IAO SB RAS, i.e., in Akademgorodok (eastern suburb of Tomsk) and in the reference station located near the Kireevsk village (forest zone, 60 km from Tomsk) at areas of about $10 \cdot 10$ m^2. To estimate the aerosol deposition flows, snow was sampled in Akademgorodok in January and April of 2006 and in the Kireevsk village only in April. The samples taken in January characterize total levels of aerosol pollution for the period from November to January, and those obtained in April – for the whole winter season.

The sampling and preparation of snow samples were conducted with regard for methodical recommendations from Refs. 4, 5, and 6. The samples were taken out of open test pits through the total thickness of snow cover, except for the 5-cm layer above the soil. The open test pit area was measured; and time (of day) from the beginning of snowfall was fixed. After snow melting at room temperature, the snow-melted water was filtered, and the solid sediment was obtained, dried, sieved to the fraction of under 1 mm, and weighted.[7]

The mineral-matter composition of the snow solid sediment was studied with use of the stereoscopic binocular microscope (SBM-9) and X-ray structure analysis at the Department of Geoecology and Geochemistry of the Tomsk Polytechnic University. Owing to the SBM-9 instrument, the percentage ratio was established for all natural and technogenic particles due to the method of comparison[8] with reference circles. The X-ray structure analysis allows determining the qualitative mineral content in samples.

All the samples of snow solid sediment were subjected to the instrumental neutron activation analysis on the content (concentration, mg/kg) of 23 chemical elements in the Laboratory of Nuclear Geochemical Investigation Methods.

The instrumental measurement data were used for calculation[5] of the aerosol flow parameters. The measured dust mass in a snow sample allows determining the dust load P_n in mg/(m$^2 \cdot$ day) or

0235-6880/07/06 474-06 $02.00

kg/(km^2 · day). According to the data of snow sampling, similar parameters (load) of environmental pollution by individual chemical elements were also calculated. At calculations, the total mass of pollutant flows, i.e., P_n, kg/(km^2 · day) and concentration of individual elements (mg/kg) in power snow were taken into account. The product of these parameters allows calculating the total load P_{total}, kg/ (km^2 · day), produced by the presence of each of these chemical elements ino the environment.[5]

As an anomaly index in the content of chemical elements, the concentration factor K_c was used, which was calculated as ratio of some element concentration in the investigated object to its mean background content, typical for the given region.[5] The K_c magnitude was calculated relative to the background values, obtained earlier for Tomsk Region and presented in Refs. 2 and 3.

When analyzing the results of snow measurements, we also used the data on aerosol content in the surface atmosphere for the period from November, 2005 to April, 2006 obtained at the aerosol station of IAO SB RAS. The round-the-clock hourly measurements of the directed aerosol scattering factor for the solid basis of submicron particles at $\lambda = 0.52$ μm (nephelometer) and mass soot concentration in the air (aethalometer) have been conducted in the automatic mode at the aerosol station located in the Akademgorodok, starting from 1996. The current data of the station are available in Internet (http://aerosol1.iao.ru).[9] These measurements give an important information on time characteristics of the submicron aerosol and soot in the surface air for the given time interval.

According to the data on the directed aerosol scattering factor, the mass concentration of submicron aerosol was estimated (due to the single parameter model of atmospheric fogs[10] for density of particles of 1.5 g/cm^3). Note that soot in the atmospheric aerosol composition, especially in winter period, is mainly formed anthropogenically from combustion of hydrocarbon fuel (fuel-energy complex, industry enterprises, transport, etc.) and in some extent characterizes the level of technogenic load on the environment. The analyzed submicron aerosol has a large lifetime in the atmosphere (several weeks) and, accordingly, is transferred by air masses at large distances (thousands of kilometers), participating on a regional scale in sedimentation on soil, water surface, snow and ice covers.

Note that results of the round-the-clock measurements of aerosol and soot concentrations, which were carried out in winter–spring period of 2001 at the aerosol station of IAO and at Kireevsk village have shown that data for these two measurement points well-correlated and more than 70% of realizations were statistically indiscernible, i.e., the anthropogenic effect of the town was manifested only in 20–30% of day realizations.[11] This allows considering that measurements at the Aerosol Station of IAO are close to background by the level of aerosol concentrations, and informative relative to the dynamics of the mid-regional background aerosol state.

Results

The results of dust load calculation show that for the Akademgorodok samples, this parameter varies within 20–40 kg/(km^2 · day) in January and 20–30 kg/(km^2 · day) in April, whereas in the Kireevsk, it varies from 16 to 20 kg/(km^2 · day) (April). The obtained values of dust load correspond to rather low pollution level [less than 250 kg/(km^2 · day)], in accordance with gradation offered in Ref. 5. However, in comparison with the ambient load for the Tomsk Region of 6 kg/(km^2 · day) by the data of Ref. 2, the four-fold excess is marked for Akademgorodok and the double one – for the Kireevsk village.

According to the SBM-9-investigation results of mineral-matter composition of the snow solid sediment samples, 14 types of particles were discovered, among them, 6 types correspond to natural formations (different types of quartz, feldspar, biogenic particles, etc), and eight types (soot, slag, metallic microspherules, mullite, etc.) – to technogenic ones. A group of particles of technogenic origin (60–70%) dominates over the group of natural particles (30–40%) in samples taken in Akademgorodok in January and April. The main fraction of pollutions referring to technogenic formations (soot, slag, mullit) are typical for the emissions of the Tomsk State District Power Station fuel and energy complex. In Kireevsk samples, the group of natural particles (70–75%) prevails over the group of particles of technogenic origin (25–30%). The major fraction of natural formations falls on quartz particles.

The results of X-ray structural analysis in samples of snow solid sediment have shown that quartz is predominantly fixed in Akademgorodok.

Tables 1 and 2 present the results of distribution of heavy metals, rare, rare-earth elements, and radioactive elements according to the data of instrumental neutron activation analysis in the areas under investigation. It should be noted that comparison of results obtained for Akademgorodok and the Kireevsk village was conducted by the April data.

Heavy metals (As, Cr, Ba, Sr, Co)

In samples of snow solid sediment, the content of Co, Sb, Ba, and Sr exceeds the background by an order of magnitude. In Akademgorodok, the maximal concentration of Ba, Co, and Sr is observed as compared to those of the Kireevsk village. However, the content of Sb and Cr is higher in snow samples from the Kireevsk village. In snow samples of Akademgorodok, the concentration of heavy metals taken in January is 1.5–2 times higher, than concentration of these metals, taken in April (see Table 1).

Table 1. Content of heavy metals and rare elements in the snow solid sediment (mg/kg) and the total load value

Index	As	Co	Sb	Cr	Ba	Sr	Rb	Cs	Hf	Ta
	Akademgorodok, January, 2006									
Mean	0.3	23.7	8.1	94.9	1290.0	280.0	58.0	5.0	6.2	1.2
Standard error	0	1.5	0.3	3.3	101.2	180.0	5.7	0.6	0.3	0.1
Minimum	0.25	21.7	7.6	90.8	1110	100	50	4.2	5.8	1
Maximum	0.25	26.7	8.6	101.5	1460	640	69	6.2	6.8	1.3
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total}, g/(km^2·day)	0.01	0.7	0.2	2.8	38.9	8.5	1.7	0.1	0.2	0.03
	Akademgorodok, April, 2006									
Mean	0.3	19.3	7.7	81.5	1030.0	100.0	49.7	3.4	5.2	1.0
Standard error	0	2.8	1.1	8.5	80.0	0	1.9	0.2	0.2	0.1
Minimum	0.25	13.9	5.7	65.6	950	100	46	3.1	4.8	0.9
Maximum	0.25	23.3	9.3	94.8	1190	100	52	3.7	5.6	1.2
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total}, g/(km^2·day)	0.01	0.5	0.2	2.2	26.8	2.6	1.3	0.1	0.1	0.03
	Kireevsk village, April, 2006									
Mean	0.3	13.5	8.9	102.2	716.7	100.0	49.0	3.8	4.7	0.7
Standard error	0	0.9	1.4	17.8	54.6	0	5.7	0.6	0.5	0.1
Minimum	0.25	12.2	6.8	83.4	610	100	38.0	3.0	4.1	0.6
Maximum	0.25	15.3	12	137.8	790	100	57.0	5.0	5.7	0.8
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total}, g/(km^2·day)	0.005	0.3	0.2	1.9	13.2	1.8	0.9	0.1	0.1	0.01
"Background,"* mg/kg	0.5	10	2.3	110	100	100	55	3.5	2.2	0.1
"Background,"* g/(km^2·day)	0.003	0.1	0.01	0.7	0.6	0.6	0.3	0.02	0.01	0.001

* Data[2] with additions from Ref. 3.

The total load produced by entering heavy metals on the environment, exceeds 2–3 times the background both in Akademgorodok and in the Kireevsk village. The value of heavy metal load is less in samples of the Kireevsk village than in those taken in Akademgorodok. In April, we observed the total load decrease in Akademgorodok by 1.5–2 times as compared to January (see Table 1).

Rare elements (Hf, Cs, Rb, Ta)

In samples of snow solid sediment, the content of rare elements 1.5–2 times exceeds the background. It should be noted that the concentration of these elements in samples of Akademgorodok and the Kireevsk village differ a little. In samples taken in Akademgorodok in January, the content of rare elements is 1.5–2 times higher, than in April (see Table 1).

Rare-earth elements (Sm, Lu, Yb, La, Sc, Tb)

The content of rare-earth elements in samples of snow solid sediment in the investigated areas 2–3 times exceeds the background.

The content of rare-earth elements in samples of Akademgorodok and the Kireevsk village differs a little. The concentrations of the given elements in samples taken in Akademgorodok in January and April also differ insignificantly (Table 2).

The total load produced by the rare-earth elements on the environment, 1.5–2 times exceeds the background both in Akademgorodok, and in the Kireevsk village. The total load in the Kireevsk village is less than in Akademgorodok. It should be noted that in samples taken in Akademgorodok in April, almost double total load decrease is observed as compared to the samples obtained in January (Table 2).

According to Ref. 2, the ratios between single elements and their groups are used as indicators for revealing the technogenic sources. The most informative features are (La/Yb), (Th/U), and ((La + Ce)/(Yb + Lu)). It was revealed that values of La/Yb and (La + Ce)/(Yb + Lu) in samples of investigation sites are close to each other (Table 3). It should be noted that La/Yb and (La + Ce)/(Yb + Lu) in samples of snow solid sediment well agree with ratios obtained in the earlier conducted investigations[3,12] near the Tomsk SDPS-2 and enterprises of nuclear-fuel cycle[3] (Table 3). It can be assumed that the main source of rare-earth elements is the fuel and energy complex.

Radioactive elements

The content of U and Th significantly exceeds the background in all analyzed samples. The concentration of radioactive elements in samples taken in Akademgorodok exceeds a little their content in samples from the Kireevsk village. The content of these elements in samples taken in Akademgorodok in January and April, differ insignificantly (see Table 2).

Table 2. Content of rare-earth and radioactive elements in the snow solid sediment (mg/kg) and the total load quantity on the snow cover

Index	Lu	La	Ce	Sm	Eu	Tb	Sc	Yb	U	Th
	Akademgorodok, January, 2006									
Mean	0.4	38.9	75.5	5.8	1.7	1.0	12.3	3.2	4.6	11.4
Standard error	0.03	1.0	5.1	0.3	0.1	0.1	0.3	0.2	0.9	1.9
Minimum	0.39	37.1	68	5.3	1.6	0.8	11.8	2.9	3.6	9.1
Maximum	0.48	40.7	85	6.1	1.8	1.2	12.7	3.6	6.4	15.1
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total}, g/(km$^2\cdot$day)	0.01	1.2	2.2	0.2	0.1	0.03	0.4	0.1	0.1	0.3
	Akademgorodok, April, 2006									
Mean	0.4	35.1	64.2	6.0	1.2	0.8	10.2	2.9	4.5	8.5
Standard error	0.02	3.2	4.9	0.7	0.1	0.1	1.0	0.3	0.4	0.4
Minimum	0.37	29.4	55	4.8	1.1	0.66	8.2	2.6	3.8	7.8
Maximum	0.44	40.6	70	7.3	1.3	0.99	11.2	3.5	5.2	9.3
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total} g/(km$^2\cdot$day)	0.01	0.9	1.7	0.2	0.03	0.02	0.3	0.08	0.1	0.2
	Kireevsk village, April, 2006									
Mean	0.4	30.6	52.1	5.1	1.0	0.7	9.0	2.4	3.1	6.8
Standard error	0.01	1.7	1.2	0.6	0.1	0.1	0.4	0.1	0.4	0.2
Minimum	0.4	27.3	50.5	4.3	0.9	0.7	8.5	2.2	2.4	6.4
Maximum	0.4	32.3	54.4	6.2	1.1	0.8	9.8	2.7	3.5	7.2
Number of samples	3	3	3	3	3	3	3	3	3	3
P_{total} g/(km$^2\cdot$day)	0.01	0.6	1.0	0.1	0.02	0.01	0.2	0.04	0.1	0.1
FEC*, mg/kg	0.2	23.7	57.2	4.8	1.3	0.67	10.2	1.96	3.5	7.8
NFC**, mg/kg	3.7	25.6	78.2	6.3	2.5	1.12	16.7	2.3	3.2	11.4
"Background,"*** mg/kg	0.1	2.8	10	0.6	1.1	0.06	7.1	0.2	0.2	2.9
"Background,"*** g/(km$^2\cdot$day)	0.0005	0.02	0.1	0.03	0.007	0.0004	0.04	0.001	0.001	0.02

* Fuel and energy complex, according to Refs. 3,12.
** Region with enterprises of nuclear-fuel cycle according to Ref. 3.
*** According to the data from Ref.2 with additions from Ref. 3.

The total load produced by radioactive elements, appreciably increase the background values in all samples. The lower total load values are observed in samples taken in the Kireevsk village, than in Akademgorodok (see Table 2). There are some differences in samples taken in January and April (see Table 2).

Table 3. Values of the relation Th/U, La/Yb, and (La+Ce)/(Yb+Lu)

Investigation areas	Th/U	La/Yb	(La+Ce)/(Yb+Lu)
Akademgorodok, January	2.5	12	31.2
Akademgorodok, April	1.9	12	29.7
Kireevsk village, April	2.1	12.6	29.2
FEC*	2.2	12.1	37.5
NFC**	3.6	11.1	17.3

See the note to Table 2.

Besides, it was revealed that content of U, Th, and Th/U in samples of the above-mentioned points well agrees with that near the Tomsk SDPS-2 (see Table 2).[3,12] It should be noted that aerosols of the studied regions has a well-defined uranium specialization (Th/U < 3).

According to the results presented in Tables 1 and 2, the geochemical associative series are constructed by the concentration factor decrease relative to the background[2,3] for Akademgorodok: U_{22}–Yb_{14}–Tb_{13}–La_{12}–Sm_{11}–$Ba_{10.3}$–$Ta_{10.2}$ – Ce_6 – $Lu_{5.4}$ – $Na_{5.2}$ – Ag_4 – $Sb_{3.3}$ – Th_3 – $Hf_{2.4}$ – $Br_{2.3}$ – $Co_{1.9}$ – $Fe_{1.8}$; for Kireevsk village: U_{16}–Yb_{12}–$Tb_{11.8}$–La_{11}–Sm_9–$Ta_{7.2}$–$Ba_{7.2}$–$Na_{5.4}$–$Lu_{5.4}$–Ce_5–Ag_4–$Sb_{3.9}$–$Au_{3.3}$–Br_3–$Th_{2.3}$–Hf_2–$Co_{1.3}$.

To compare the investigation results, we have applied the cluster analysis aimed at division of the set of chemical elements into groups. The elements with the highest similarity of the paired Pearson[13] correlation coefficients r are united into these groups.

As follows from Fig. 1, the geochemical spectra of elements in the snow solid sediment differ.

In samples taken in Akademgorodok in January, the following close association connections are distinguished: iron–barium, samarium–natrium, bromine–strontium, calcium–cobalt, and uranium–chromium. In samples taken there in April, the associations of rare-earth elements are distinguished (lanthanum–lutecium, europium–samarium), bromine–rubidium, ytterbium–barium and noble metals with radioactive elements. In its turn, in samples taken in the Kireevsk village, the samarium–

antimony, europium–barium, and natrium–cobalt (Fig. 1) associations are marked.

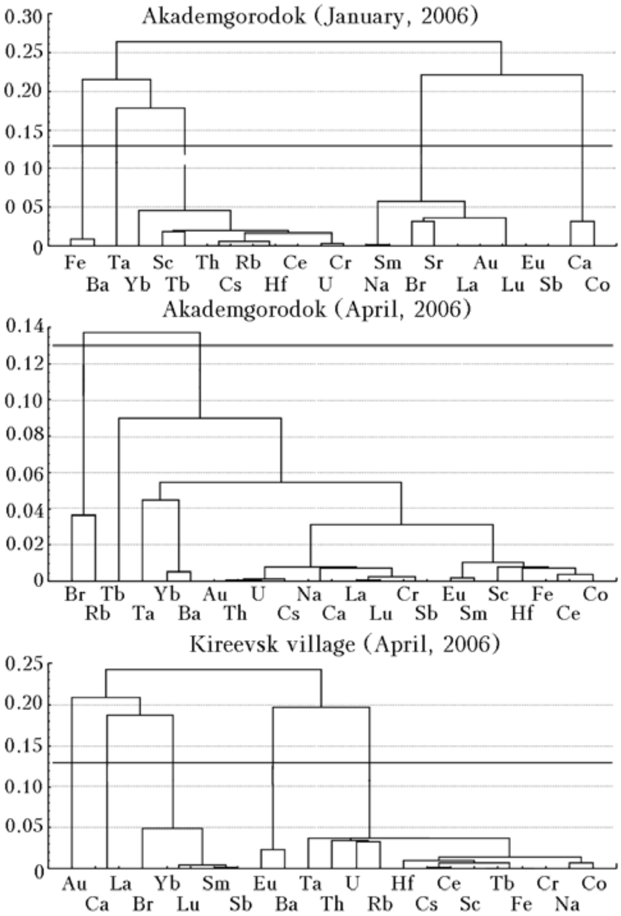

Fig. 1. The correlation matrix dendrograms of the geochemical spectrum of snow solid sediment. Along the *Y*-axis, *r* equal to 0.13 is laid off.

Time variability of submicron aerosol and soot concentrations

Figure 2 illustrates the time behavior of average daily concentrations of solid basis of the submicron aerosol and soot by the measurements at the Aerosol Station of IAO in the surface layer for the period corresponding to the time of pollution accumulation in the analyzed snow samples, i.e., from November, 2005 to April, 2006.

The presented time pattern of aerosol concentrations corresponds to the earlier established typical pattern of annual behavior of the aerosol and soot concentrations in the surface layer by the results of long-term measurements.[14] The annual behavior of these aerosol characteristics are specified by the well-defined winter maximum and summer minimum. The marked form of annual behaviors of the aerosol and soot concentrations is natural and has a property of inter-annual stability. As follows from Fig. 2, where the data are given with the 30-day sliding averaging, the maximum of mean values of the aerosol and soot concentrations (about 33.6 and 3.64 $\mu g/m^3$, respectively) is observed in January in the period of snow accumulation. When passing to April, there is a gradual concentration decrease down to the corresponding values of about 18.3 and 1.26 $\mu g/m^3$.

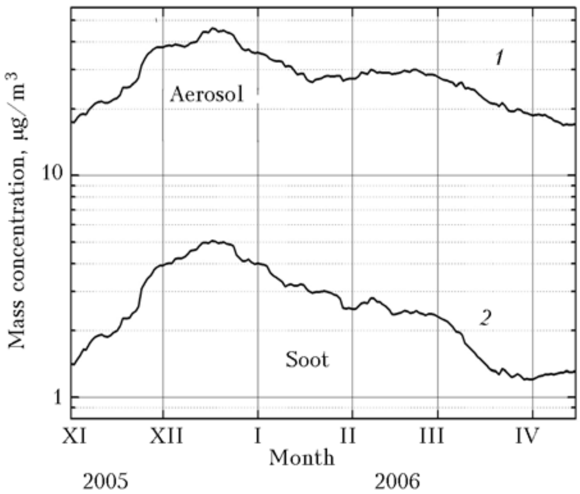

Fig. 2. Time behavior of average daily values of the submicron aerosol (*1*) and soot (*2*) concentrations.

Thus, when going from winter to spring seasons, the gradual decrease of submicron aerosol concentration approximately by 1.8 times and the more significant soot concentration decrease (by 2.9 times) are observed. A similar time variability tendency of the aerosol and soot concentrations in the surface air qualitatively agrees with the above-mentioned features of decrease of aerosol deposition flows in the snow when going from winter to spring seasons.

The coordinated dynamics of aerosol depositions in the snow cover and the concentration of aerosol characteristics in the surface layer should be explained by the seasonal variability of factors determining generation and distribution of the aerosol component in the boundary layer of the atmosphere. The increased level of aerosol and soot in the surface layer in the cold period of the year is caused by sharp intensification of anthropogenic aerosol sources (heating season) and the action of surface thermal inversions typical for this period and preventing the aerosol emissions into the higher-lying atmospheric layers. When approaching the spring period, the action of the indicated factors weakens. Besides, the seasonal temperature variations in the boundary atmosphere leads to the altitude increase of the mixing layer and the aerosol content decrease in the surface layer, therefore, to reduction of aerosol pollution of the snow cover.

Conclusion

The results of geochemical analysis have revealed that the samples of snow solid sediment in Akademgorodok have higher values of heavy metal concentrations (As, Ba, Sr, Co) and radioactive elements (U, Th) as compared to those of the Kireevsk village. In its turn, for the Kireevsk village, the higher levels of Cr and Sb are marked. The concentrations of rare-earth elements (Sm, Lu, Yb, La, Sc, Tb) and rare elements (Hf, Cs, Rb, Ta) for two measurement points differ insignificantly. The total load value of all the considered elements is by 1.5–2 times higher in Akademgorodok, than in the Kireevsk village.

It should be noted that, according to the indicator ratios of the elements La/Yb, Th/U, and (La + Ce)/(Yb + Lu), the samples taken in the Kireevsk village and in Akademgorodok, are close to each other and agree with the corresponding values for the urbanized territories (with thermal power coal-fired stations).

When analyzing the time variability of aerosol flows in Akademgorodok, a stable peculiarity of dust load value decrease, as well as total load of each individual chemical element are revealed during passing from January to April. The concentration of heavy metals and rare elements also decreases from January to April. However, the concentrations of radioactive and rare-earth elements are not subjected to significant variations.

The coordinated tendencies to the gradual decrease of aerosol deposition flows in snow, the submicron aerosol and soot concentrations in the surface air when passing to the spring season, are caused by the decrease of the intensity of polluted components at the end of the heating season (heat power complex), and the seasonal variability of temperature conditions in the boundary atmosphere.

Acknowledgements

This work was supported in part by the Russian Foundation for Basic Research (Grant No. 06-05-64393).

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