Sources and sinks of anthropogenic microelements in the Arctic atmosphere: tendencies in variations from 1981 to 2005

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We have analyzed data of long-term observations on the transport of air masses toward three sites in the Russian Arctic and backward. From this analysis we have estimated the mean atmospheric concentrations and surface fluxes of anthropogenic microelements in the considered Arctic regions for two decades (1986–1995 and 1996–2005). The analyses revealed tendencies in variation of the level of air pollution and admixture deposition rate in Russian Arctic caused by long-term variations of the processes of atmospheric circulation in different seasons. At the end of twentieth and beginning of twenty first centuries, the changes in the processes of pollution and scavenging of Arctic air are seasonally non-unique. Spring is characterized by the tendency toward reduction of the content of anthropogenic microelements in the aerosol of the central part of the Russian Arctic; the total annual fall-outs on the surface in the Arctic have also decreased. In magnitude, this effect is quite comparable with variations of concentrations and fall-outs to be expected from the decreased emission from sources in these years. From the viewpoint of arrival of pollutants from the middle latitudes to Arctic, reduction of the efficiency of atmospheric channel may led to increase of the role of river sink because of fall of anthropogenic components from the atmosphere directly to river waters, as well as to snow and soil on the territory of the river catchments yet in subarctic regions.

Introduction

Content of one or another component in the atmospheric air is determined by the balance between the powers of its sources and sinks. Sources of the anthropogenic components of the atmosphere are industrial enterprises, cities, or regions where many enterprises and even whole industrial complexes are concentrated. Influence of these territories on the atmospheric composition in remote regions of the globe strongly depends on the processes of circulation of air masses, which determine the direction and range of pollutant transport in the atmosphere. On the other hand, one of the most effective sinks of the aerosol admixture from the atmosphere, in the course of the admixture transport by air masses away from a source, is deposition on the surface. Efficiency of this process depends not only on the properties of air mass (humidity and precipitation), but also on the quality of the surface, which, to a certain extent, depends on the air circulation, determining pathways (trajectories) of air mass over the territories and surfaces.

The changes of the environmental climatic characteristics of the northern hemisphere, observed at the end of twentieth and beginning of twenty first centuries,[1,2] have attracted enhanced interest to all phenomena influencing the composition and properties of natural objects on different scales. In particular, the change has also occurred in the atmospheric circulation over the Northern polar region (NPR) which, by virtue of the aforesaid, could not help influencing the sources and sinks of anthropogenic components of the atmosphere on the pollution level of air and underlying surface in Arctic.

In earlier publications,[3–5] we studied the sources and sinks of anthropogenic microelements in atmospheric aerosol of Russian Arctic, based on analysis of 10-year synoptic data for 1986–1995. The present paper continues the studies of regularities and seasonal features of transport of air mass and anthropogenic aerosol constituents to three sites located in Arctic zone on the territory of Russia. The initial data series has been extended to 2005, allowing us to consider the materials of the present study as climatically significant results, uncovering the tendencies of variations of the processes of air and anthropogenic admixture transport in the lower troposphere and the associated conditions governing atmospheric composition in Russian Arctic.

The goal of the paper was the study and estimation of long-term tendencies in variations of the following processes and characteristics:

spatial distribution of air masses arriving at Russian Arctic and leaving it in different seasons;

potentialities and influence of most significant industrial zones and regions on the composition of the Arctic atmosphere;

mean concentrations of anthropogenic microelements in the near-ground atmosphere of Arctic;

fluxes of anthropogenic microelements to the underlying surface, influence of atmospheric channel of pollutant transport on the environment of Arctic as a whole.

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Statement of the problem and method of estimation

In recent years, the trajectory analysis approach has been used not only for study of origins the components of concrete air samples, but also for investigation of mean regularities in the processes of long-range transport of air masses and substance in the atmosphere.[6–9] The reliability of the obtained results is determined to a large degree by the quality and spatial density of the meteorological information upon which the calculations are based, as well as by the methods of presentation and description of transformation of a substance during its transport in the atmosphere. Use of large trajectory arrays permits calculation of admixture transport along each trajectory (taking into account the corresponding admixture sources and the processes of admixture scavenging) by summing their contributions and analyzing the spatial admixture distribution field.[7,8]

Other approach involves the statistical processing of the spatial distribution of the trajectories themselves and subsequent use of the averaged parameters for determination of sources[9] or estimation of admixture spread from concrete large source regions. Precisely this variant, suggested[10] and extensively described[3,4,11] by ourselves earlier, was used in this study in order to leave unchanged the method of analysis of all long-term variations. We will now give brief account of the most critical points of this approach.

Observation sites for our studies are three points in the Russian Arctic, located in Franz Joseph Land (FJL; 81.1°N, 56.3°E), on archipelago Severnaya Zemlya (SZ; 79.5°N, 95.4°E), and on Vrangel island (VR; 71.0°N, 178.5°W). For each of these points we analyzed the 5-day forward (air out of the site) and backward (air toward the site) trajectories of air motion for each day in one of the four months for 20-year period (1986–2005). The trajectories were calculated using data of Hydrometeorologic Center of Russia for 0000 GMT (with the step of 6 h) on isobaric surfaces 925 and 850 hPa for January, April, July, and October. Considering these months as representative of the corresponding seasons, we can gain idea of the seasonal variations of the studied processes and characteristics.

For each of the sites, the forward and backward trajectories (separately) were classified according to their spatial position.[10] We calculated the frequencies of arrival of air masses to observation sites from continents, Arctic, and Pacific and Atlantic Oceans, as well as analogous distributions of the leaving air for the four seasons of the year. The term "Arctic" in our classification means a region poleward of 70°N plus the remaining, more southern part of Greenland.

Most extensively and consistently, the method of quantitative estimates of admixture transport within the framework of this approach has been described by Vinogradova and Ponomareva[4] and by Vinogradova and Egorov.[10] Solution of the balance equation of admixture mass in air flow during admixture transport from the source i to the point j can be presented by the following formula for the admixture concentration C_{ij} at the observation point j:

$$C_{ij} = Q_i(1 - \alpha_i)Z_{ij}, \qquad (1)$$

where Q_i is the power of emissions of admixture from the source i; $\alpha_i \approx 0.1$ is the fraction of the admixture, deposited on the ground near the source i. The function Z_{ij}, we termed the transport efficiency function (TEF), characterizes all atmospheric processes, influencing the admixture transport on its way from the source i to the point j. It depends on the probability and velocity of air motion, which are determined from the trajectories, as well as on the rate of admixture deposition from the atmosphere during transport, that depends on the conditions of vertical mixing and precipitation. The TEF calculations use empirical data on spatiotemporal variations of temperature inversions in the near-ground tropospheric layer[12,13] and statistical characteristics of clouds[14] and precipitation.[15] Thus, in the framework of this approach, TEF values are used to estimate the potentialities of one or another source region in affecting the atmospheric composition at the considered sites.

We stress that our approach and all subsequent considerations and conclusions pertain to the so-called conservative admixture, passive with respect to the chemical conversions in air during transport, or to separate microelements whose identification is possible irrespective of the concrete chemical composition of aerosol substance.

The rate of admixture deposition on the underlying surface is presented as the sum of dry deposition and scavenging by precipitation. Values of the dry deposition rate for different seasons were chosen according to the available literature data (experiment and model estimates), while the rates of deposition by precipitation were calculated using data of long-term observations on the atmospheric precipitation from Ref. 15. The total rate of deposition on the surface was assumed constant for each season and site, independent of admixture type, and invariable during admixture transport to the observation site. Thus, all the results discussed below are valid for atmospheric constituents whose rates of deposition on the surface in different seasons are equal to 0.05–0.07 in winter, 0.1–0.2 in spring, 0.9–1.2 in summer, and 0.4–0.8 cm $\cdot$ s^{-1} in fall.

The process of scavenging of the Arctic atmosphere from admixtures was considered as a sum of three processes: 1) dry deposition, 2) deposition by precipitation on the underlying surface inside the region (poleward of 70°N), and 3) atmospheric advection out of the region to more southern latitudes. Having known the distribution of air masses, leaving the observation sites, as well as the rate of admixture deposition on the underlying surface, we can estimate the balance of conservative admixture after it leaves the observation sites.[5,16]

In cold months, the rate of admixture deposition on the surface in Arctic is very low, and a marked admixture fraction (30–50%) stays in the Arctic

atmosphere for longer than 5 days and mixes in this region like in a huge reservoir. Then, assuming that the air, arriving at the observation sites from Arctic (regions poleward of 70°N), is as polluted as the air at the observation site, and neglecting the contribution of marine air to the abundances of the anthropogenic components, the resulting average (during month) concentration C_j of the admixture in the region of observation site j can be estimated by the equation

$$C_j = \Sigma C_{ij} + q_j C_j, \quad (2)$$

where q_j is the frequency of arrival of the Arctic air masses to the site j, and the summation is performed over all sources i. The contribution of the Arctic air, i.e., the magnitude of the second term in right-hand side of formula (2), is considerable (25–50%).[4,11] It is somewhat larger in spring than in winter, and it is a bit lower on VR than in FJL and SZ. In calculations of the summer and fall concentrations, the contribution of Arctic air can be disregarded since in the warm half-year, the deposition rates of anthropogenic admixtures are an order of magnitude larger than in the cold period of the year.

Having known the average admixture concentration C_j in the near-ground mixing layer H_j and the rate of admixture deposition on the surface K_j for each site j, we can estimate the mean fall-outs of this admixture per unit surface:

$$D_j = C_j K_j H_j, \quad (3)$$

where H_j is weighted average height of the mixing layer, determined from the data on height and frequency of occurrence of near-ground and elevated temperature inversion layers near the observation site j.

The method of estimation of the mass of admixture, blown by airflows form Arctic, on the basis of statistics of forward trajectories of air mass transport has been described extensively in Refs. 16 and 17.

In this paper, the estimates of atmospheric admixture concentrations and admixture fall-outs on the surface are presented for four chemical elements, namely As, Ni, Pb, and Cd of predominately anthropogenic origin, whose sources are different industrial and domestic processes. The literature[18,19] provides data on the powers and composition of atmospheric emissions of the largest European and Russian industrial regions, considered to be the sources of these elements in the atmospheric aerosol. To determine the effect, associated precisely with the change of circulation processes in the atmosphere, the powers of emissions of the considered microelements from the source regions are kept fixed at the level according to these studies dated in early 1980s. Change of the emission of sources in the last years of the twentieth century and possible consequences of these changes will be considered briefly in the last section.

Spatial distributions of air mass transport trajectories

Strong interannual variability of the spatial distributions of air masses, arriving at the observation sites or leaving them, is caused by variations of the atmospheric pressure field and atmospheric circulation regimes. Figure 1 presents the variations of the spatial distributions of air masses, arriving at FJL, SZ, and VR from the territory of Europe, Asia, America, Arctic, and Pacific Ocean in April during 25 years from 1981 to 2005 (with addition of data for 1981–1985 from Ref. 10). It is seen that FJL and SZ in early 1980s were characterized by about equally frequent arrivals of air from Europe, Asia, and Arctic; while at the beginning of twenty first century, there was a marked prevalence of arctic air masses with a reduction of the rate of arrival of air from Eurasia.

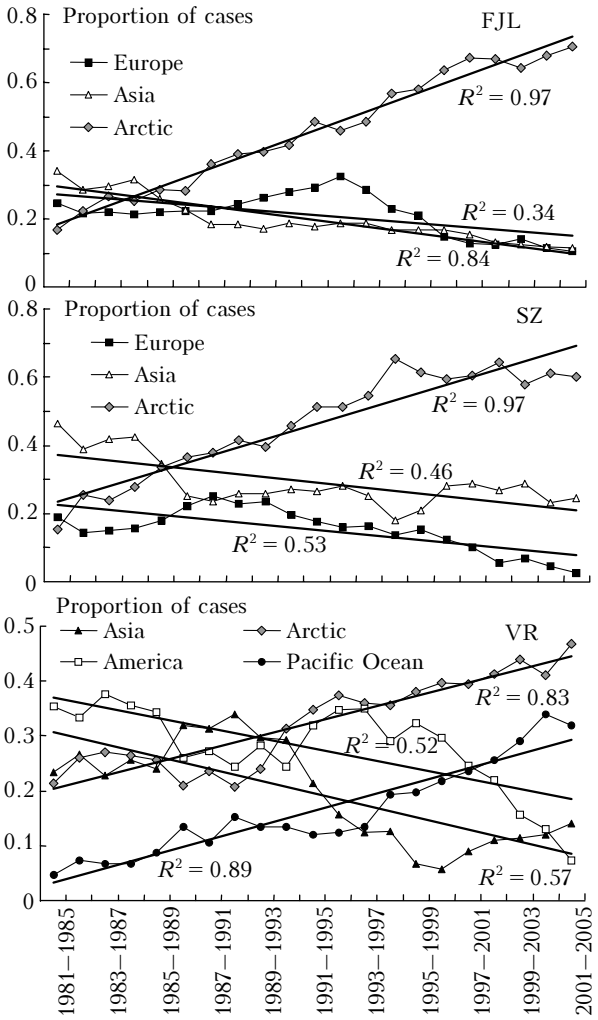

Fig. 1. Variations of the spatial distributions of air masses arriving at FJL, SZ, and VR from the territories of Europe, Asia, America, Arctic, and Pacific Ocean in April. Shown are the five-year average frequencies of arrival. Straight lines show linear fit, R^2 is the confidence factor.

For VR, the total pattern is somewhat more complicated, though certain distinct regularities are present. Whereas at the beginning of the analyzed period, VR was dominated by air from the territory of America with almost no air supply from Pacific; at the end of the period, the air arrived most frequently from Arctic and from the territory of Pacific Ocean,

while the fraction of continental air masses has considerably decreased. By analyzing quantitatively these distributions for each month, we can conclude that, in last 20 years of the twentieth century, the atmospheric circulation in the northern polar region has considerably changed, and that a number of statistical regularities exist in the studied distributions.

Table 1 shows the tendencies revealed in variations (over period 1986–2005) of the spatial distributions of air masses arriving at the observation sites in different seasons. The estimates of the linear trend were made using 5-year smoothed time dependences of the frequencies of arrival of air masses. For clarity of presentation, the frequency variations (over the entire period) are expressed in percent of their values for the first five years (1986–1990). It is seen that, at all sites, in arriving air masses there was increasing fraction of Arctic air. Conversely, the fraction of air coming to FJL and SZ from Europe and to VR from America and Asia decreased. We revealed no variations in distributions for October in FJL and SZ and for January in SZ, when the considered trends were found to be very small and/or insignificant.

As to the spatial distribution of trajectories along which the air leaves the observation sites, almost no significant linear trends in these distributions for the studied years were detected. It can only be stated with confidence that in October, the air from VR had more frequently left to the territory of Pacific Ocean and more rarely to Arctic and Asia. Possibly, this agrees with data from Ref. 20, indicating increase of southern meridional component of wind velocity at the latitude 70°N near 180°E at the end of twentieth and beginning of twenty-first centuries compared with the preceding 30 years.

Table 1. Characteristics of the linear approximation of 5-year smoothed frequencies of arrival of air from Europe, Asia, Arctic, America, and Pacific Ocean. Entries are trend (in percent of values for 1986–1990)/confidence factor of the linear approximation R^2

Site	Air arrival from	January	April	July	October
FJL	Europe	–23/0.33	**–71/0.80**	–54/0.38	–
	Asia	–	–	–	–
	Arctic	**+41/0.78**	**+75/0.89**	+33/0.47	–
SZ	Europe	–	**–85/0.95**	–56/0.55	–
	Asia	–	–	**–73/0.81**	–
	Arctic	–	+26/0.57	+81/0.45	–
VR	Asia	–68/0.45	–84/0.69	–51/0.60	–75/0.68
	Arctic	+112/0.40	**+61/0.79**	**+174/0.71**	**+307/0.72**
	America	–28/0.36	–	–43/0.64	**–61/0.78**
	Pacific Ocean	–	+123/0.60	–	–

Note. Dashes correspond to small values of trend or R^2. Numbers in bold are for trends corresponding to $R^2 > 0.7$.

Tables 2 and 3 present the correlation coefficients, showing the interrelations of different pathways of air arrival at the observation sites. For instance, in FJL and SZ, the air from Europe competes with air masses from Arctic; while on SZ in January there occurs competition between air masses from Europe and Asia (see Table 2). Quite a different situation is characteristic of Vrangel Island, where the Asian air competes with air masses from Arctic or America in the presence of one powerful channel more from Pacific water basin.

Table 2. Correlation coefficients (calculated from annual data) of the frequencies of arrival of air masses at FJL, SZ, and VR from the territories of Europe (*Eur*), Asia (*A*), Arctic (*Arc*), America (*Am*), and Pacific Ocean (*PO*) or escape of air to these territories in different months

Site of arrival/escape	Month	Arrival of air from territories of			Escape of air to territories		
		Eur–A	*Eur–Arc*	*A–Arc*	*Eur–A*	*Eur–Arc*	*A–Arc*
FJL	January	0.44	–0.47	–0.35	–0.14	–0.23	0.19
	April	0.24	**–0.86**	–0.47	–0.49	0.11	–0.10
	July	0.01	**–0.82**	–0.14	–0.20	–0.23	–0.41
	October	–0.31	**–0.72**	–0.33	–0.31	–0.22	–0.26
SZ	January	**–0.61**	–0.46	–0.36	–0.32	–0.25	–0.09
	April	–0.27	**–0.53**	**–0.56**	**–0.60**	–0.057	0.21
	July	–0.16	**–0.68**	–0.46	–0.01	–0.02	–0.46
	October	–0.31	**–0.72**	–0.33	–0.31	–0.22	–0.27
VR		*A–Am*	*A–Arc*	*Am–Arc*	*A–Am*	*A–Arc*	*Am–Arc*
	January	–0.18	**–0.57**	–0.49	**–0.52**	–0.39	–0.18
	April	–0.49	–0.26	–0.29	–0.29	–0.32	–0.31
	July	–0.16	**–0.65**	–0.23	**–0.53**	–0.25	**–0.51**
	October	0.11	**–0.69**	**–0.58**	–0.04	–0.34	–0.21
		A–PO	*Am–PO*	*PO–Arc*	*A–PO*	*Am–PO*	*PO–Arc*
	January	0.13	–0.15	–0.44	0.07	–0.23	**–0.69**
	April	–0.04	–0.44	–0.41	–0.17	–0.44	–0.40
	July	–0.04	–0.31	–0.49	–0.24	0.046	–0.44
	October	–0.33	–0.14	–0.18	–0.21	–0.20	**–0.75**

Note. Numbers in bold are larger than 0.5 in absolute value.

Table 3. Correlation coefficients (calculated from annual data) of frequencies of arrival of air masses from same territory (Europe, Asia, or Arctic) at two of the three considered sites

Air arrival from	Month	FJL–SZ	FJL–VR	СЗ–ВР
Europe	January	**0.60**	–	–
	April	**0.69**	–	–
	July	0.47	–	–
	October	**0.76**	–	–
Asia	January	**0.54**	–0.11	–0.04
	April	0.34	0.06	–0.25
	July	0.37	0.13	**0.59**
	October	**0.64**	–0.15	–0.26
Arctic	January	0.41	–0.22	0.08
	April	0.45	0.18	–0.18
	July	0.39	0.31	0.41
	October	**0.53**	0.001	–0.07

N o t e . Numbers in bold exceed 0.5 in absolute value. Dashes correspond to cases when the correlation coefficient was not calculated because almost no air from Europe arrives at VR.

As a result (see Table 3), in FJL and SZ the frequencies of arrival of European air in different years well correlate with each other, for air masses from Asia this is characteristic only in January and October, and for Arctic air this is the case only in October.

Thus, FJL and SZ frequently fall within a single large-scale atmospheric system (cyclone or anticyclone). The air circulation regimes in FJL and VR regions are not statistically and actually linked with each other. For the SZ/VR pair this link is apparent only in July, when the frequencies of arrival of air masses from Asia and Arctic correlate with each other.

Thus, the variations of atmospheric circulation for the considered 20 years were such that on the Russian coast of Arctic the zonality of air motion has increased at the expense of meridional transport. From the viewpoint of pollution transport from more southern latitudes to Arctic, these tendencies must reduce the effectiveness of pollution of the Arctic atmosphere. This will be shown in sections below.

Pollution sources and their contributions

The continental air arriving at Arctic may carry anthropogenic pollutions as a result of passage of air masses over big industrial regions. The locations of all big industrial regions of the Northern Hemisphere, which were considered as possible sources of anthropogenic pollution of the atmosphere at the observation sites, is presented schematically, e.g., in Refs. 11 and 17.

References 3, 4, and 11 provide information on the numerical values of TEF (see comments below formula (1)) for a series of source regions over which the studied trajectories passed in each season in the period 1986–1995. The changes of TEF values in 1996–2005 compared to the preceding decades for most significant sources are shown in Fig. 2.

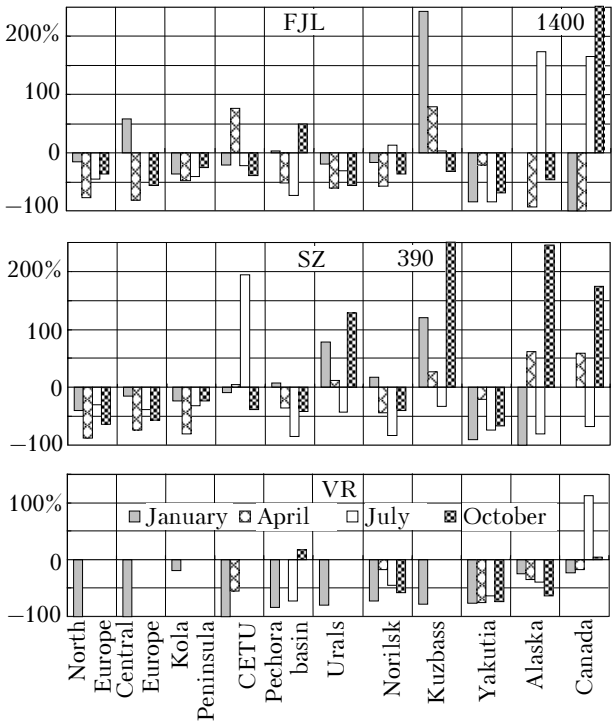

Fig. 2. Relative variations of efficiency function of admixture transport from source regions to FJL, SZ, and VR in different months. Plotted are differences of the average values for the periods 1996–2005 and 1986–1995, scaled to the value in earlier period.

It is seen that for the considered 20 years, the potentialities of most source regions in influencing the air composition in Russian Arctic have decreased. Opposite tendencies are marked mainly on SZ and, to a lesser degree, in FJL. It is worthy to note the increase of efficiency function of transport from sources located on the territory of North America (Alaska and Canada), as well as from remote Russian regions, namely Kuzbass and Urals.

Note that TEF characterizes only the possibility of each source to emit pollutants to the atmosphere near the corresponding site. At the same time, the contribution of each region to the abundance of concrete admixtures is determined by formula (1) using data on atmospheric emissions Q_i of these admixtures from the considered regions. Quite possibly, most significant sources may have been not those with larger TEF, and not those with more powerful emissions.

Contributions of source regions as well as atmospheric concentrations and fall-out of microelements on the underlying surface are estimated only for FJL and SZ, and not for VR because of the absence of sufficiently complete data on composition and powers of atmospheric emissions by industrial regions located in North America.

References 4 and 11 contain information on the interrelation of contributions of source regions to the

concentration of the considered microelements in FJL and SZ, characteristic for the period 1986–1995 when most significant sources of the considered microelements were North Europe, Kola Peninsula, Norilsk, and Urals, as well as the Arctic air itself in cold half-year [see formula (2)]. Variations of the contributions of source regions to abundance of anthropogenic microelements in the atmosphere of FJL and SZ in different seasons, revealed in 1996–2005, compared with variations in preceding decades, are non-unique (Table 4).

Table 4. Change (for the decade 1996–2005 compared to 1986–1995) of the contributions of source regions to atmospheric pollution in FJL and SZ in different months

Source region	January		April		July		October	
	FJL	SZ	FJL	SZ	FJL	SZ	FJL	SZ
North Europe	–	–	– –	–	–	+		–
Kola Peninsula	–		–	–		+ +	+	
Urals	–	+	–	+		+		+ +
Norilsk			–		+	– –		– –
Kuzbass	+	+	+	+				+ +
Arctic	+	–	+ +	+				

Note. Plus/minus symbols mean increase/decrease by more than 5%; double plus/minus symbols mean more than 20% change; and empty cells mean less than 5% change.

There has been greater contribution of Ural Region, especially to SZ atmosphere, primarily in terms of arsenic and lead, while Kuzbass has been a considerable source of all considered elements to the atmosphere of both sites. In April, there was a considerable increase of the contribution of the Arctic air itself as a source of pollutants accumulated in the cold months. However, the contributions of North Europe and Norilsk markedly decreased. As a result, in recent years the level of these microelements in the atmosphere in FJL and SZ is mainly determined by six sources, whose contributions in different months are on the average interrelated as they are in Fig. 3.

In analysis of the seasonal differences in these distributions it is necessary to keep in mind that the absolute concentration of anthropogenic admixture in summer is approximately an order of magnitude lower than in winter.

For VR, a still valid conclusion[4,11] is that on the average in all seasons the contribution of North American sources of anthropogenic pollution to the composition of the atmosphere of East Arctic predominates (over Eurasian sources).

Atmospheric concentrations of microelements

Vinogradova[4] and Vinogradova and Ponomareva[11] showed that the estimates, based on the proposed method of the average atmospheric concentrations of the considered microelements in the Russian Arctic for the period 1986–1995, well correspond to real long-term array of experimental data, obtained on the territory of North Canada in 1981–1995,[21] and examples of data measured in Severnaya Zemlya in late 1980s.[22] In the framework of this study, we compared average (monthly mean) near-ground concentrations of microelements, calculated for the period 1996–2005, with the results for the preceding decade. Figure 4 presents relative variations of concentrations of arsenic, nickel, lead, and cadmium in the atmosphere over FJL and SZ in different months.

It is seen that in the FJL atmosphere at the end of the twenties and beginning of twenty-first century, all the considered elements (with the exception of Cd in winter) decreased in content, whereas for SZ the pattern is not that unique. In SZ, the concentrations grew in winter (for all elements) and fall (with the exception of As and Ni) with more considerable (than for FJL) decrease of concentrations in spring and summer (because of the increase of the amplitude of seasonal behavior).

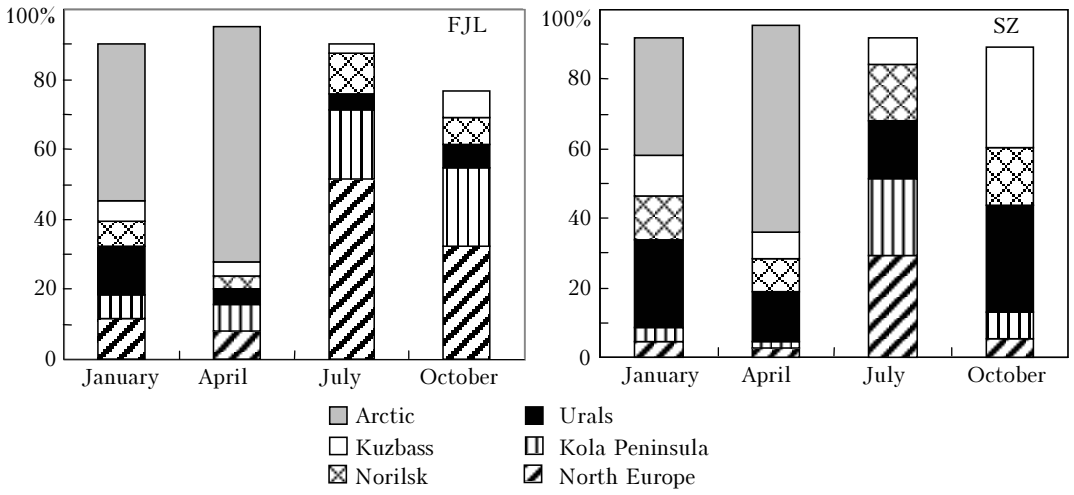

Fig. 3. Relative contributions of six most significant source regions to the admixture abundance in the atmosphere over FJL and SZ in different months. Plotted are average ratios for all considered microelements.

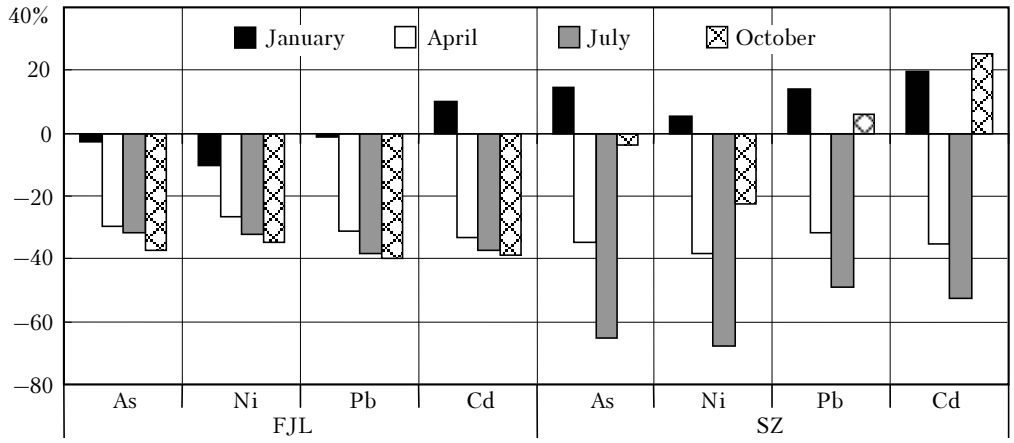

Fig. 4. Relative variations of concentrations of arsenic, nickel, lead, and cadmium in the atmosphere over FJL and SZ in different months. Plotted is the difference of average concentrations for the periods 1996–2005 and 1986–1995, scaled to the value for earlier period.

No long-term continuous observations of elemental composition of atmospheric aerosol in Russian Arctic were performed; therefore, we compared the tendencies in variations of the anthropogenic microelement concentration in the atmosphere over Russian Arctic, determined by ourselves, with observed trends of concentrations of anthropogenic aerosol components, obtained in Canadian Arctic[21] and Alaska[23] (Fig. 5).

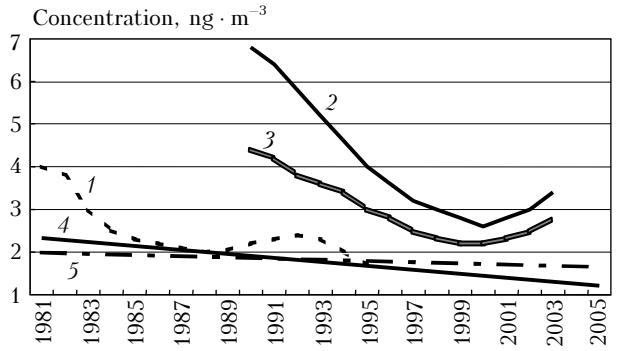

Fig. 5. Comparison of calculated tendencies in variations of lead concentration in archipelago Severnaya Zemlya (for April (solid straight line *4*) and January + April (dashed-dotted line *5*) with the experimental results, obtained for lead (curve *1*) and soot (curve *2*) in Canadian Arctic and Alaska (curve *3*)[20,21] in cold season of the year.

Indeed, at the end of twentieth century, there is a tendency toward reduction of pollution of the Arctic atmosphere, which in the real atmosphere is caused not only by change of circulation, but also by the decrease of emission of sources,[24,25] to be discussed in the last section.

Scavenging of the Arctic atmosphere

The fall-outs of anthropogenic (frequently toxic) admixtures from the atmosphere on the underlying surface play an important role in the present life and evolution of Arctic ecosystems. Our estimates showed that, for the considered 20 years, the interrelation between vertical (deposition on the surface) and horizontal (advection to latitudes southward of 70°N) mechanisms of scavenging of the Arctic atmosphere did not change qualitatively, and the quantitative differences do not exceed the calculation error.

In the cold half-year, the amounts of admixtures, brought from Arctic by airflows, deposited on the surface, and staying in the atmosphere poleward of 70°N are approximately the same. In the warm half-year, almost all admixtures, carried to Arctic, deposited within 5 days on the underlying surface in the region poleward of 70°N. As a result, the Arctic atmosphere is scavenged in different seasons as follows: 1) in winter the advection of admixture by airflows out of Arctic is most effective; 2) in spring, this process and the process of dry deposition on the surface have comparable efficiencies; 3) in cold half-year, the admixture deposition by precipitation is insignificant; and 4) in warm half-year, the Arctic atmosphere is scavenged predominately by admixture deposition on the surface, mostly due to washing out by precipitation.

Figure 6 shows changes that took place in 1996–2005 (compared with the preceding years) in powers of the processes scavenging the atmosphere of Arctic from anthropogenic admixtures in different months.

It can be stated for sure that fall-outs of all elements on the underlying surface decrease in April, July, and annually and increase in January. The average fluxes of the four considered microelements per unit area in FJL and SZ regions for period 1996–2005 are presented in Table 5.

The difference between the two observation sites is 4 to 7%. From Table 5 we see that the anthropogenic admixture flux onto the surface in Arctic has seasonal dependence, with maximum at the warm time of the year. Assuming that all the four seasons last 3 months each, we can estimate that, via atmospheric channel, Arctic receives yearly about 140 t of arsenic, 440 t of nickel, 1600 t of lead, and 40 t of cadmium. This is (depending on the concrete

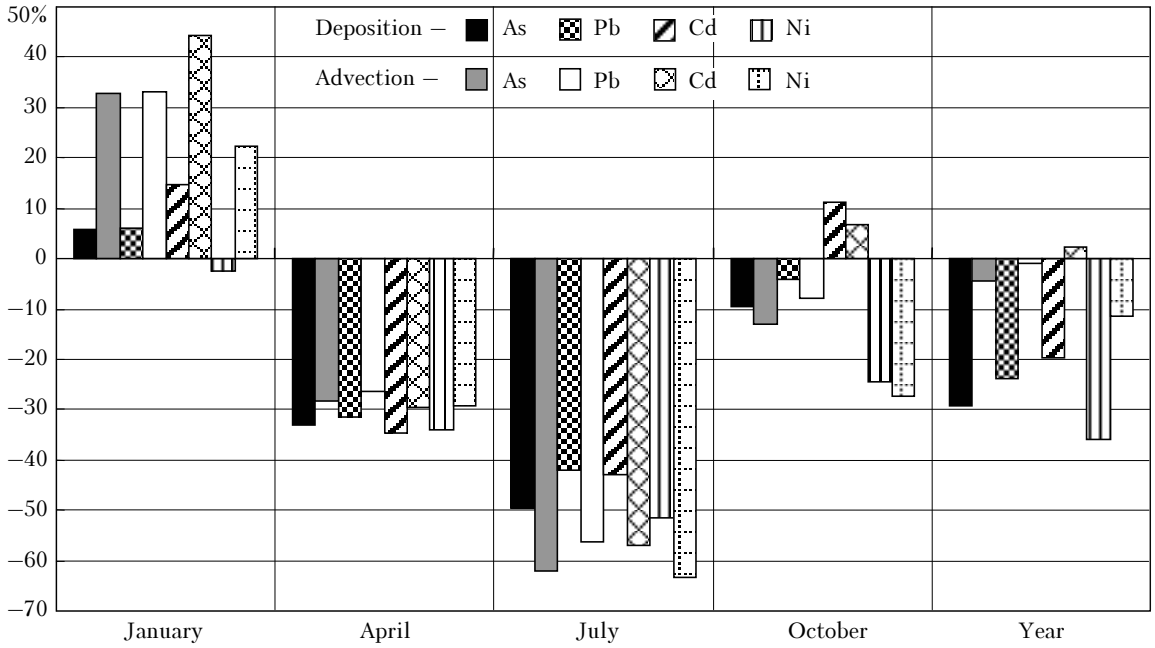

Fig. 6. Relative variations of the powers of the vertical (deposition) and horizontal (advection) atmospheric scavenging channels. Plotted is the difference in the amount of substance, removed from the atmosphere monthly/yearly, for the periods 1996–2005 and 1986–1995, scaled to the value for earlier period.

element) 6–10% of the atmospheric emissions of all source regions supplying this chemical element to the considered Arctic regions, and does not exceed 1–3% of the emissions of all anthropogenic sources of Europe and Russian part of Asia. The amount of anthropogenic microelements arriving in Arctic in 1996–2005 has been less than in the preceding decade by 15% for Cd and by 30% for Ni. Note that the absolute vertical fluxes from the atmosphere to the surface, presented in Table 5, are only the anthropogenic part of the corresponding quantities, without the contribution from natural sources (sea surface, soil, volcanoes, etc.) to the atmospheric abundances and, hence, to fall-outs of the considered elements.

Table 5. Average flux of anthropogenic chemical elements from the atmosphere to the underlying surface in the region of FJL and SZ in 1996–2005, $\mu g \cdot m^{-2} \cdot month^{-1}$ ($g \cdot km^{-2} \cdot month^{-1}$)

Month	As	Ni	Pb	Cd
January	0.29	0.92	3.5	0.08
April	0.29	0.94	3.4	0.08
July	0.66	2.20	7.9	0.19
October	0.85	2.46	9.2	0.26

As was already noted above, in the Arctic atmosphere there take place mixing and redistribution of anthropogenic pollutants in the Northern Hemisphere. Thus, on the average, about half of the anthropogenic admixtures brought by airflows to Arctic during 1986–1995, was deposited on the surface, and the other half was blown by airflows back to southern latitudes. However, in the last decade this balance has changed. Now, only about 1/3 of the admixtures brought by air masses during a year form anthropogenic sources to Arctic is blown to south, and the remaining 2/3 of the admixtures fall on the underlying surface poleward of 70°N and may participate subsequently in biochemical cycles of the Arctic ecological systems.

Possible influence of other factors

The main factor, determining the sources of anthropogenic admixtures in the Arctic atmosphere, is the reduction of the powers of emissions of practically all considered microelements to the atmosphere that took place in 1990s.[24] From Meteorological Synthesizing Center "Vostok" (MSC-V)[25] data on variations of lead and cadmium emitted in these years by European countries we can conclude that in Europe the lead emissions have decreased on the average by a factor of 5, while cadmium emissions by a factor of 2 (both emissions by a factor of 1.5 in Russian part). Had the circulation processes been invariable, this would lead to a proportional decrease of concentrations of these elements in the Arctic atmosphere. Therefore, the effect of 40% decrease of concentration (see Fig. 4) as a result of change of the circulation processes in the atmosphere, obtained by ourselves, is quite comparable with the consequences which can be caused by changes of emissions from source regions. Moreover, both these factors reduce the pollution of the Arctic atmosphere, leading to experimentally observed dependences such as those exhibited by lead and soot concentrations presented in Fig. 5.

The factor influencing the admixture sinks from the atmosphere is the amount of precipitation. Data from Ref. 2 show that at the end of the twentieth century the precipitation amount has increased on the territory of the 60–70°N belt (especially in the

Eurasian sector) and decreased at latitudes to the North from it (70–80°). This has to increase the rate of admixture deposition on the underlying surface along the pathway of transport to Arctic (in regions equatorwards of 70°N) and, hence, to decrease the content of anthropogenic pollutants in the atmosphere of the Arctic latitudes. A consequence may be an increase of the role of river sink as a channel of pollutant arrival to the Arctic environment since already in subpolar regions, the atmospheric pollutants will fall directly to river waters as well as on snow, ice, and soil on the territory of river catchment basins.

Conclusions

Trajectory analysis of air mass transport to the Russian Arctic has shown that during 25 years (from 1981 to 2005) zonality in the processes of air circulation over the territory of North Eurasia prevailed over the meridional transport, especially in spring.

As a result of changes in the circulation processes, the variations of the atmospheric concentrations of As, Ni, Pb, and Cd and their fall-outs on the underlying surface in Russian Arctic are non-unique: they decrease all year in FJL, and they decrease only in spring and summer and increase in winter on Severnaya Zemlya. However, the total annual fall-outs have decreased by 20–35% (depending on particular microelement).

During a year, Arctic receives about 6–10% of the atmospheric emissions of the considered microelements released by all source regions from which these elements are delivered to the regions poleward of 70°N that does not exceed 1–3% of the emissions of all anthropogenic sources of Europe and Russian part of Asia. In 1996–2005, the arrival of anthropogenic microelements in Arctic has been 15–30% lower than the amount brought there in the preceding decade.

In cold seasons, in the Arctic atmosphere there take place mixing and redistribution of anthropogenic pollutants whose sources are located mainly in midlatitudes of the Northern Hemisphere. In the last decade, on the average, about 1/3 of admixtures, brought by air masses to Arctic from anthropogenic sources, are blown back to southern latitudes. The remaining 2/3 of the admixtures fall on the underlying surface poleward of 70° and may participate subsequently in biogeochemical cycles of Arctic ecosystems.

Decrease of the efficiency of air channel as a supplier of anthropogenic admixtures from midlatitudes to Arctic may lead to an increase of the role of river sink in this process because in subpolar regions the pollutants from the atmosphere will settle down to river waters, as well on snow, ice, and soil on the territory of river catchment basins.

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