Fifteen-year trends in processes of air mass and anthropogenic aerosol transport over the Laptev Sea region

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Fifteen-year (1986–2000) daily 5-day forth and back trajectories of air mass motion for one month (January, April, July, and October) in each season are analyzed for a geographic point located roughly at the center of the Laptev Sea. The seasonal behavior of mean atmospheric concentrations and surface deposition of six anthropogenic chemical elements (As, Ni, Pb, V, Zn, Cd) is calculated. The 15-year variations of the studied characteristics are estimated as linear trends of 8-year sliding averages. It is shown that for 15 years the role of European regions in pollution of the Laptev Sea has decreased, while the role of Asian regions has, to the contrary, increased. As a result of variation of only atmospheric circulation processes, the summer and spring pollution levels almost did not change, while the trends of winter and fall levels were almost identical, but with the opposite signs (increase in winter and decrease in fall). The annual mean fallouts of the considered aerosol microelements on the surface of the Laptev Sea decreased by no more than 20%. The actual decrease of atmospheric emissions of anthropogenic pollutants that occurred in Eurasia in these years should further enhance this effect.

Synoptic and meteorological features of the Arctic atmosphere are such that in the cold season (winter and spring) it contains marked amounts of anthropogenic components transported to the Arctic from mid-latitudinal industrial regions.[1] These components fall on the surface and pollute snow, ice, and water of the Arctic Ocean (AO). For some microelements (for example, lead) the atmospheric contribution to the composition of water in the central part of AO is most strong.[2] The Laptev Sea is the region, where arctic ice, melting in the Fram Strait and the Greenland Sea, begins its circumpolar drift. Therefore, the chemical composition of the Laptev Sea may affect the composition and properties of water in the central part of AO and northern Atlantic. Our previous paper[3] dealt with the study of the effect of atmospheric aerosol on the mean pollution level of air and water in the Laptev Sea in the period from 1986 to 1995. Recent observations[4,5] showed that variations of a number of geophysical parameters (air temperature, precipitation, permafrost boundary, etc.) occurred at the verge of the third millennium are indicative of climatic shifts in the Northern Hemisphere. These phenomena should undoubtedly have effect on the processes determining the level and seasonal variations of atmospheric pollution in the Arctic. The objective of this work was to study changes in atmospheric circulation that occurred in the central part of the Russian Arctic for 15 years (since 1986 until 2000), as well as their effect on the processes of transport and surface deposition of anthropogenic aerosols.

Methods and approaches

For the geographic site with the coordinates 77°N, 125°E located roughly at the center of the Laptev Sea (Fig. 1), we analyzed 5-day forth and back air motion trajectories calculated in the Russian Hydrology and Meteorology Committee from 925 and 850 hPa isobaric surfaces as for 00:00 GMT (with the interval of 6 h) for every day of January, April, July, and October since 1986 until 2000. Assuming these months representative of the corresponding seasons, we can believe the mean regularities in the studied process in particular month to be characteristic of the whole season.

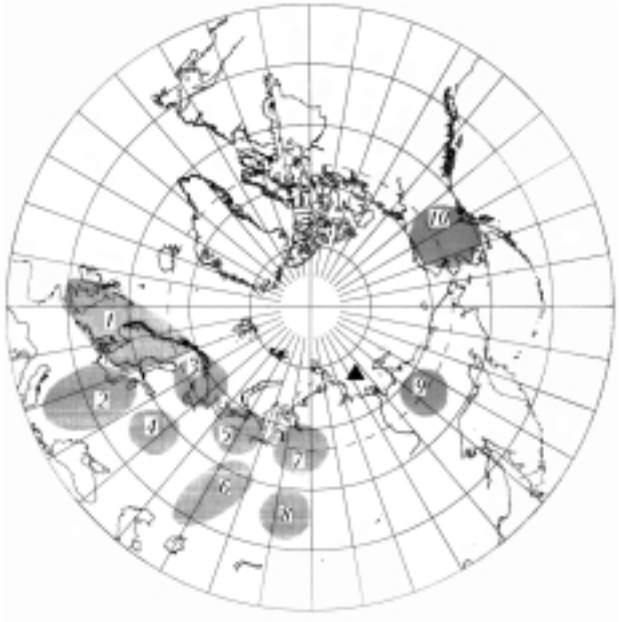

Fig. 1. Schematic map of large industrial regions being the sources of atmospheric pollution in the Laptev Sea region: Northern Europe *1*, Central Europe *2*, Kola Peninsula *3*, central part of the European territory of the former USSR (CPET) *4*, Pechora basin *5*, Ural *6*, Norilsk region *7*, Kuzbass *8*, Yakutiya *9*, Alaska *10*.

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For each month, all the forth and back trajectories were divided into groups by their spatial features: where air masses moving by these trajectories are from and where they are going to.[6,7] All the ambient territory was divided into the following parts: continents (Europe, Asia and America), the Arctic (area inside 70°N circle plus the rest more southern part of Greenland) and oceans (Pacific and Atlantic Oceans separately). As was noted in Ref. 3, in all the seasons the interannual variability of atmospheric circulation in the Laptev Sea region is very wide. Therefore, to increase the reliability of the results, we first calculated 5-year sliding average of the frequency of occurrence for trajectories of the corresponding group and then used them to determine the linear trend of this frequency in time for the considered years.

The technique used for estimation of mean atmospheric concentrations of aerosol trace components and their flows onto the surface that is based on the data of statistical analysis of large arrays of air motion trajectories for Arctic regions is described thoroughly in Ref. 8. This technique is applicable to chemically inactive atmospheric components, whose surface deposition rates can be believed equal to: 0.05–0.08 $cm \cdot s^{-1}$ in winter, 0.1–0.2 $cm \cdot s^{-1}$ in spring, 0.9–1.2 $cm \cdot s^{-1}$ in summer, and 0.4–0.8 $cm \cdot s^{-1}$ in fall. Examples of such components are anthropogenic heavy metals, as well as arsenic and elemental carbon (soot).

As was shown in Ref. 3, ten large industrial regions of the Northern Hemisphere are the sources of atmospheric pollution in the Laptev Sea region (Fig. 1).

For each region, we can estimate the potential possibility (different in different seasons) of affecting the atmospheric pollution level at the observation site. For this purpose, the so-called transport efficiency function (TEF) is introduced. The value of this function for each source region depends on the probability and the speed of air motion between this source and the observation site. The TEF value is also determined by the conditions of vertical mixing in the atmospheric surface layer and the rate of deposition onto the surface for the time of transport. The equations and values of the parameters for TEF calculation were given in Refs. 3 and 8. Based on the data of many-year observations, they account for seasonal differences in characteristics of temperature inversions, cloudiness and precipitation in the central part of the Russian Arctic. Within the framework of this approach, the mean atmospheric concentration of a trace element C at the observation site is calculated for each season as a sum of contributions coming from the continental C^{cont} and arctic C^{Arc} air:

$$C = C^{cont} + C^{Arc} = \sum C_i + qC, \qquad (1)$$

where C_i is the mean concentration produced by the region i, which is proportional to the corresponding TEF and the amount of atmospheric emissions in this region; q is the mean frequency of air income from the Arctic for winter or spring (for summer and fall $C^{Arc} = 0$). As was shown in Ref. 8, the air in the Arctic and its components are actively mixing in winter and spring. Therefore, the second term in Eq. (1) accounts for pollution of the Arctic atmosphere itself on the assumption of the uniform distribution of a trace element all over the Arctic. In summer and fall, this effect is absent since the rate of element deposition onto the surface is roughly an order of magnitude higher than in other seasons.

The mean flow of the anthropogenic component onto the surface was calculated, as in Refs. 7 and 8, namely, from the mean atmospheric concentration of this component using the results of statistical analysis of the distributions of forth trajectories of air mass transport from the observation site.

Taking into account the wide year-to-year variability in atmospheric circulation processes, to obtain the representative statistical characteristics of the trajectories between the source regions and the observation site, averaging over the sufficiently large number of years is necessary in calculation of TEF. For example, in Refs. 3 and 8 averaging was carried out over 10-year arrays of trajectories. In this case, when our goal was to reveal the temporal trends of TEF variations for 15 years, averaging was carried out for each season over 8-year moving sets of trajectories. Then we calculated 8-year TEF values and then the 15-year TEF trends using a linear approximation. The atmospheric concentration of an element and its surface flow were determined for each season separately from the initial (1986–1993) and final (1993–2000) sets of TEF values determined from regression straight lines. The amounts emitted by the source regions were fixed at the values given in Refs. 9 and 10, which allowed us to analyze the effect of only variations in atmospheric processes on the level of air pollution in the Arctic region under consideration.

Air mass transport

The results of analysis of the spatial distributions of forth and back trajectories are depicted in Fig. 2 for each season in the form of two histograms characterizing the circulation processes in the first and last five years of the 15-year period under consideration. These results were obtained by linear approximation of the time dependence of 5-year sliding averages of the corresponding frequencies. It can be seen that, in general, the air exchange between the analyzed Arctic region and neighboring territories has changed significantly for the last 15 years. From our data, we can distinguish the following characteristic features in variations of the air exchange process over the Laptev Sea region since 1986 until 2000: (1) in all seasons, the air after passage over the observation site more and more often stayed in the Arctic and rarely went to Asia; (2) in all seasons, except for summer, the air more and more rarely came to the observation site from Europe and more often from Asia; (3) the frequency of air income from the Arctic decreased in winter and fall and increased in spring; (4) in winter and spring the marine air, which first came from the Atlantic, by the end of the studied 15-year period began to come from the territory of the Pacific Ocean. Thus, for the considered 15-year period, the western transport in the central part of the Russian Arctic has gradually changed to the eastern one.

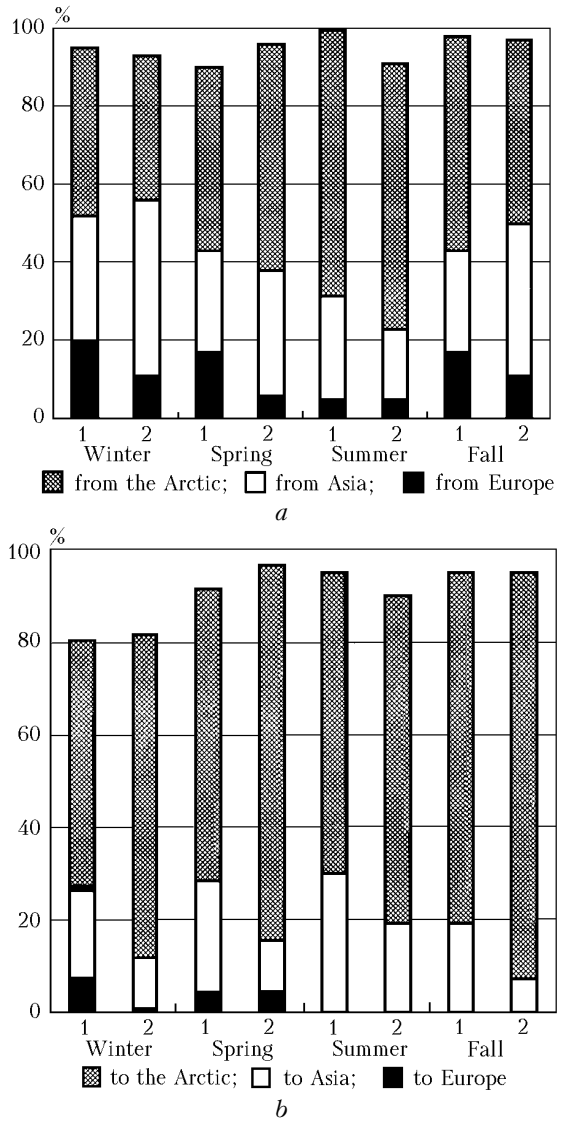

Fig. 2. Variation of the frequency of air income from different territories (*a*) and the frequencies of air going off to different trajectories (*b*) in different seasons from 1986 to 2000. The limiting results of linear approximation of 5-year averages: periods of 1986–1990 (*1*) and 1996–2000 (*2*). Missing (up to 100%) fractions are associated with air masses coming from the American and oceanic territories and going off to these territories.

Transport of anthropogenic microelements on aerosol particles

Figure 3 depicts the 8-year average TEF values for the most significant source regions in different seasons, as well as linear trends of their variations in the period considered. Trends for summer were not calculated because of poor statistics (accumulated for eight years) for individual sources. It can be seen that the decrease in the fraction of the European air and, to the contrary, the increase in the fraction of Asian air for the 15 years considered (Fig. 2) are qualitatively reflected, to some degree, in variations of the potential capability of European and Asian regions to affect atmospheric pollution over the observation site. Thus, for example, the TEF values for the Kola Peninsula and Northern Europe decreased, while for Kuzbass and Yakutiya they increased in all seasons (Fig. 3). The trends of variation of TEF values for such sources as Norilsk region, Ural, and Pechora basin that are located near the border between Europe and Asia have the opposite signs in different seasons.

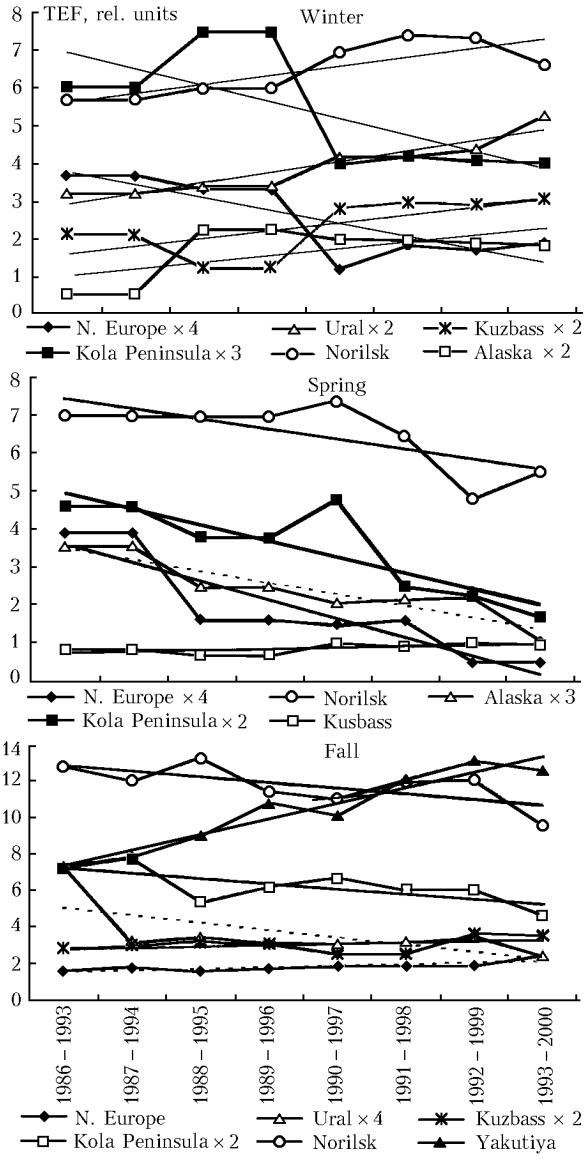

Fig. 3. Variation of TEF for most significant source regions in different seasons. Linear trends of 8-year averages.

Consider now the effect of the above changes in the air mass transport processes and the TEF values of the source regions on the level of atmospheric pollution over the Laptev Sea region with six anthropogenic microelements (As, Ni, Pb, V, Zn, Cd) that are of primary importance from the viewpoint of their negative effect on the environment and human health.[11]

Table 1 summarizes the information about 15-year variation of the average atmospheric concentrations of these six microelements at the observation site and their average fluxes onto the surface of the Laptev Sea in different seasons as calculated by Eq. (1) taking into account the data of Refs. 9 and 10 on the composition and emission rates of the Eurasian source regions. Figure 4 shows seasonal variations of the average atmospheric concentration of a trace aerosol element and its flux onto the surface of the Laptev Sea (662 000 km^2) for lead taken as an example. The qualitatively similar histograms can be drawn for the rest five microelements as well. Thus, for 15 years the average atmospheric concentrations of the anthropogenic microelements almost did not change in spring, while in winter and fall their relative variations were almost identical in value, but opposite in sign (increase in winter and decrease in fall). Some quantitative difference in the changes of the flux onto the surface, as compared with the corresponding changes of the atmospheric concentrations, is connected with variations of the conditions of clearing of the Arctic atmosphere in this period. As was noted earlier, air masses, having once passed through the observation site, more and more often remained in the Arctic, not going beyond the 70°N latitude (see Fig. 2). As a result, in the process of clearing of the Arctic atmosphere, the role of the "vertical" mechanism of the pollutant deposition onto the surface increased as compared with its "horizontal" removal by air flows from the Arctic (for more details see Ref. 12).

Table 1. Variations of average atmospheric concentrations of microelements and their average flows onto the surface of the Laptev Sea (662 000 km^2) from 1986–1993 (beginning) to 1993–2000 (end)

Element	Winter		Spring		Summer	Fall	
	Beginning	End	Beginning	End	Average	Beginning	End
	Concentration, $ng \cdot m^{-3}$						
As	0.13	0.17	0.14	0.12	0.019	0.16	0.12
Ni	0.41	0.50	0.47	0.39	0.074	0.51	0.39
Pb	1.2	1.6	0.86	0.86	0.075	1.35	1.0
V	0.30	0.40	0.17	0.16	0.014	0.33	0.22
Zn	0.73	1.1	0.60	0.66	0.023	0.80	0.70
Cd	0.030	0.042	0.028	0.027	0.0024	0.035	0.029
Change in the concentration	Increases 1.3 times		Slightly decreases		–	Decreases 1.3 times	
	Flows, $kg \cdot month^{-1}$						
As	70	100	145	130	60	350	260
Ni	220	300	480	430	230	1100	870
Pb	630	950	870	940	240	3000	2200
V	150	220	170	170	44	730	490
Zn	390	640	610	720	73	1800	1500
Cd	17	26	28	30	7.3	77	64
Change in the flow	Increases 1.5 times		Slightly varies		–	Decreases 1.3 times	

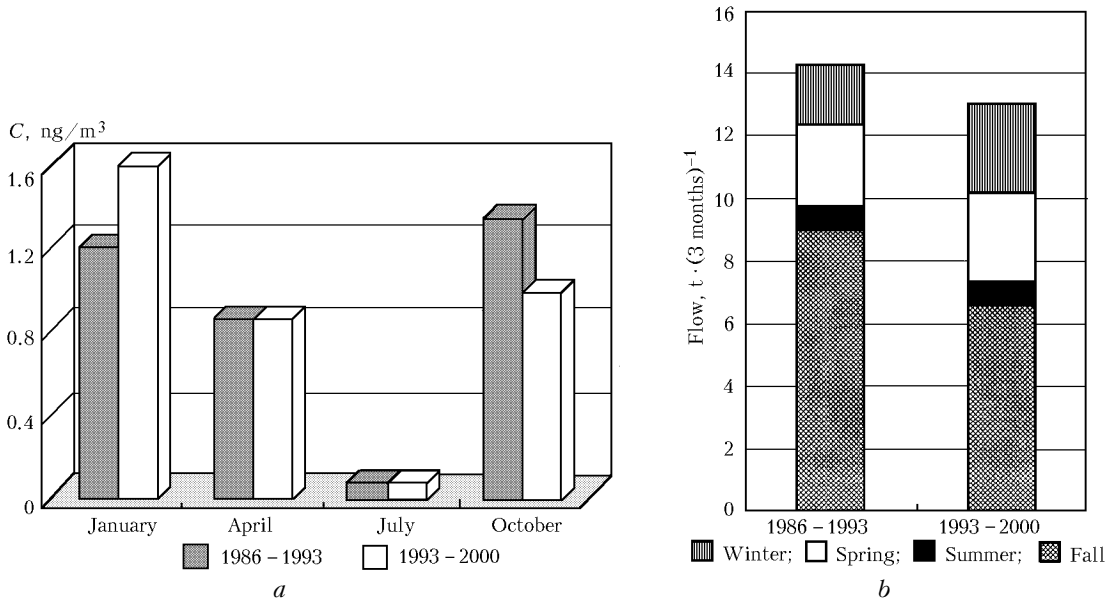

Fig. 4. Seasonal variations of the average atmospheric concentration of lead (*a*) and its deposition (for 3 months in each season and for a year) on the surface of the Laptev Sea (*b*).

Thus, according to our estimates, the annual mean depositions of such elements as As, Ni, Pb and V onto the surface of the Laptev Sea decreased only due to the changes in the air mass transport processes in the period from 1986 to 2000, but no more than by 20% (Table 2 and Fig. 4*b*). This was favored by seasonal differences in the absolute values of average fluxes onto the surface, namely, by the fact that the surface deposition of aerosol in the Laptev Sea region reaches its maximum in the fall (see Table 1). The annual fluxes of Cd and Zn onto the surface almost did not change, which is connected with the increase of TEF from Kuzbass region, whose atmospheric emissions contain these elements at relatively high concentrations, in all seasons. If we also take into account the decrease in the anthropogenic emissions in Europe and industrial regions of the former USSR that occurred in the 1990s (Ref. 13), then it can be stated that the fluxes of anthropogenic aerosol onto the surface of the Laptev Sea decreased in the past fifteen years of the 20th century.

Table 2. Variations in the annual fluxes (ton·year^{-1}) of microelements onto the surface of the Laptev Sea from 1986–1993 (beginning) to 1993–2000 (end)

Element	As	Ni	Pb	V	Zn	Cd
Beginning	1.9	6.1	14.2	3.3	8.6	0.39
End	1.6	5.5	13.0	2.8	8.8	0.38
Beginning/end	1.2	1.1	1.1	1.2	0.98	1.03

Conclusions

1. In the processes of air mass motion across the Laptev Sea in the period from 1986 to 2000, we can distinguish some tendencies characteristic of seasons and of the whole year: (a) in all seasons, having passed across the observation site, the air more and more often remained in the Arctic and more and more rarely went to Asia; (b) in all seasons except for summer, the air came to the observation site more and more rarely from Europe and more and more often from Asia; (c) the frequency of air income from the Arctic decreased in winter and fall and increased in spring; (d) in winter and spring the marine air, which first came from the Atlantic, by the end of the studied 15-year period began to come from the Pacific Ocean.

2. The potential possibilities of different source regions to contribute to atmospheric pollution in the Laptev Sea region changed in different ways: (a) in all seasons the effect of the Kola Peninsula and Northern Europe decreased, while those of Kuzbass and Yakutiya increased; (b) the trends of variation of the effect of such regions as Norilsk, Ural, and Pechora basin had opposite signs in different seasons.

3. Assuming that the emissions from large industrial regions in Europe and the Asian part of Russia did not change for the last 15 years from 1986 to 2000, the mean atmospheric concentrations of the six anthropogenic microelements transported on aerosol particles (As, Ni, Pb, V, Zn and Cd) and their mean fluxes onto the surface varied as follows:

– the atmospheric concentrations in spring almost did not change, while in winter and fall they increased and decreased roughly 1.3 times, and in summer the concentrations of anthropogenic microelements are so low, that their variations can hardly be estimated, and the effect is negligible.

– the flows onto the surface of the Laptev Sea increased roughly 1.5 times in winter, almost did not change in spring, and decreased roughly 1.3 times in fall, but the total annual fluxes of the studied elements onto the Laptev Sea slightly decrease for the considered 15-year period.

In conclusion, it should be emphasized once more that a decrease in the atmospheric emissions of the considered microelements that occurred in the 1990s in European and Asian part of Russia should additionally reduce the effect of aerosol pollution of the atmosphere and the surface in the central part of Russian Arctic in the end of the twentieth century.

Acknowledgments

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