Contribution of atmospheric aerosol to water pollution of the Laptev Sea

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Ten-year (1986–1995) series of forward and backward trajectories of air mass transport to the region of the Laptev Sea are analyzed. Seasonal variations in average atmospheric concentrations and flows onto the sea surface are estimated for six anthropogenic chemical elements (As, Ni, Pb, V, Zn, Cd) by modeling the air transport of pollutants to the Arctic. The main atmospheric pollution sources for the Laptev Sea basin are shown to be Norilsk, Ural, and Kuzbass regions, whose contributions vary in different seasons and for different kinds of pollutants. The average flows of anthropogenic atmospheric constituents onto the surface of the Laptev Sea are maximal in spring and fall and minimal in summer. The annually average contribution of atmospheric aerosol to anthropogenic pollution of water of the central part of the Laptev Sea can make about one third of that drained by rivers.

Recent investigations[1] have shown that the contribution of aerosol deposited from the atmosphere onto the Arctic Ocean (AO) surface can make 10% in the water suspended matter and in bottom deposits, that is, much higher than expected earlier. The arctic atmosphere in view of its synoptic and meteorological features in the cold season (winter and spring) contains a marked amount of anthropogenic components carried to the Arctic from midlatitude industrial regions.[2] As these components deposit onto the surface, they pollute snow, ice, and water of the Arctic Ocean. The Laptev Sea is a starting region for circumpolar drift of arctic ice, which melts in the Fram Strait and in the Greenland Sea. Therefore, the chemical composition of snow and ice from the Laptev Sea may affect the composition and properties of water in the central part of the Arctic Sea and in Northern Atlantic.

The aim of this work was to estimate the mean amount of typically anthropogenic atmospheric components (for example, some heavy metals) deposited yearly onto the surface of the Laptev Sea.

The composition of the atmospheric aerosol over the Laptev Sea has been studied experimentally in 1995 only in the summer–fall period (Ref. 1), because in other periods such study is rather difficult due to severe arctic climate. However, because of the wide year-to-year variability of atmospheric conditions and, consequently, the aerosol composition in the Arctic,[3,4] to obtain the reliable information about the mean levels of air pollution, accumulation of experimental data for many years is necessary. An alternative approach to solution of this problem is based on the analysis of many-year series of synoptic information and simulation of transport of pollutants to the Arctic.[5] Just such technique was used in this work.

For observations, we took a site with the coordinates 77°N, 125°E situated roughly at the center of the Laptev Sea (Fig. 1). For this site we analyzed the five-day forward (carrying away) and backward (bringing) trajectories of air motion calculated at the Hydrology and Meteorology Center of Russia from isobaric surfaces of 925 and 850 hPa at 00:00 GMT (with six-hour interval) for every day of January, April, July, and October from 1986 to 1995. Assuming that the selected months are representative for the corresponding seasons, the use of the ten-year series of initial data allows us to find the long-term mean seasonal regularities in the processes under study.

Arctic air masses are separated from midlatitude air by the so-called arctic front, which is a zone of efficient aerosol mixing and removal of pollutants from the atmosphere onto the surface. The mean position of the arctic front in winter and summer (according to the data of Ref. 6) is shown in Fig. 1, along with the spread of the southernmost points of the considered backward trajectories at the level of 850 hPa for January and July. Air inside the arctic front can move rather easily in the northern direction, thus transporting pollutants from midlatitudes to the Arctic. This is especially important in winter, when arctic air masses cover territories of many large industrial regions of the Northern Hemisphere. In summer the arctic front is significantly shifted to the north, thus preventing penetration of pollutants from most industrial regions to the Arctic.

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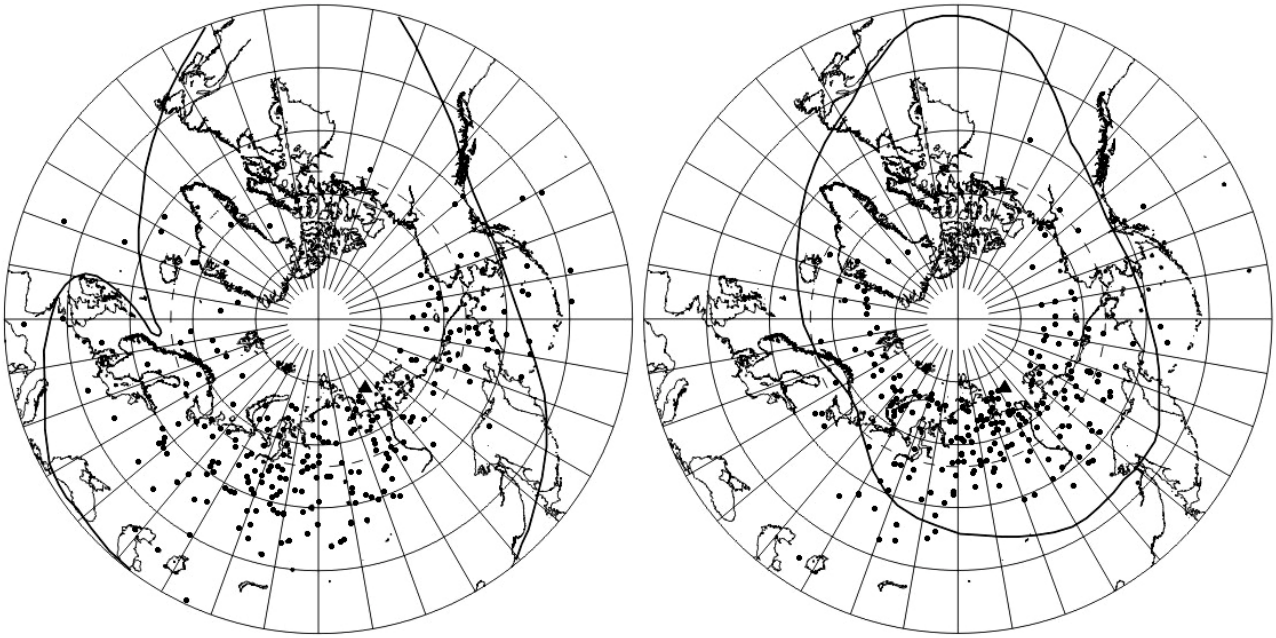

Fig. 1. Geographic location of the observation site (triangle) and the arctic front (solid lines) from Ref. 6 and the southernmost point of back trajectories calculated at the level of 850 hPa in winter (*a*) and summer (*b*).

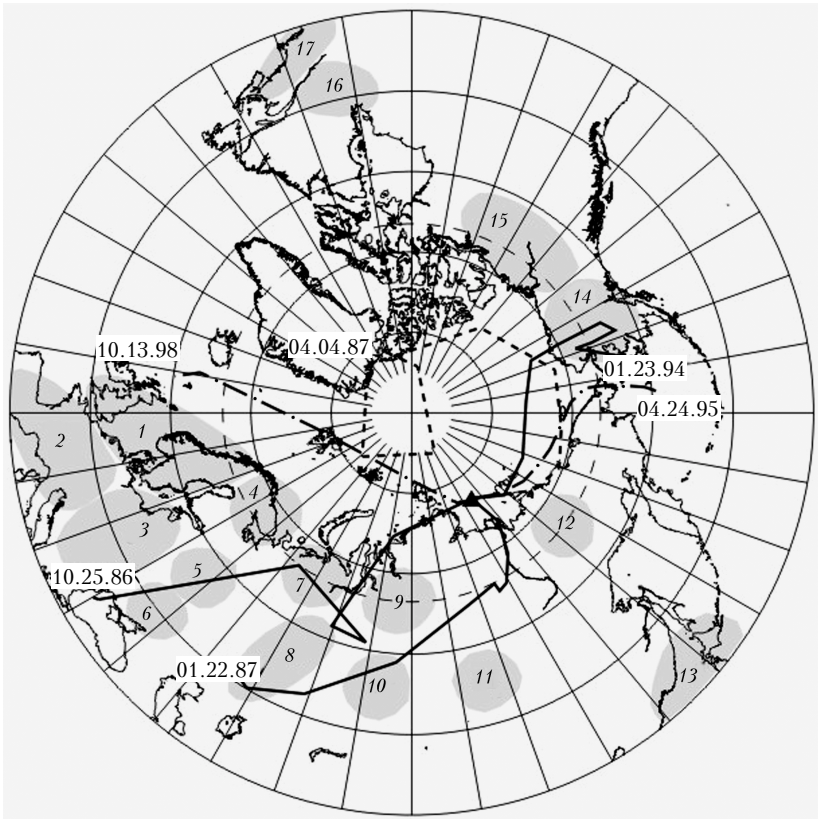

Fig. 2. Examples of trajectories at the level of 850 hPa, along which continental (solid lines), arctic (dashed lines), and marine (dot-and-dash lines) air came to the region of the Laptev Sea. Industrial regions – sources of atmospheric pollution: Northern Europe (*1*), Western Europe (*2*), Central Europe (*3*), Kola Peninsula (*4*), Central European territory (CET) of the former USSR (*5*), Donetsk (*6*), Pechora basin (*7*), Ural (*8*), Norilsk (*9*), Kuzbass (*10*), Baikal (*11*), Yakutiya (*12*), Japan (*13*), Alaska (*14*), Canada (*15*), Sudbury (*16*), US East Coast (*17*).

All forward and backward trajectories were divided into three groups depending on the directions of air mass motion along them. The technique of such division was described in detail in Refs. 5 and 7. Figure 2 exemplifies some backward trajectories, along which continental, arctic, or marine air comes to the observation site. The term "Arctic" in this paper refers to the region bounded by 70°N parallel and the adjacent more southern part of Greenland.

As was already mentioned, the year-to-year variability of atmospheric conditions is very wide in the northern polar region. To demonstrate this, Fig. 3 depicts the relation of the frequencies of occurrence of backward trajectories for air motion to the observation site from Europe, Asia, Arctic, and America in January of different years within the decade under study. It can be seen that, for instance, in 1988 the frequencies of arrival of European, Asian, and Arctic air were roughly equal, in 1990 the air came from the Arctic in almost all cases, and in 1994 the frequency of air arrival from America (which was negligibly small in all other years) was almost equal to the frequency of arrival of arctic air masses, and the European air did not come at all.

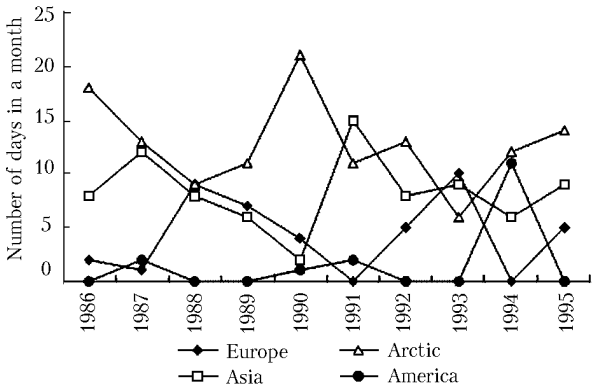

Fig. 3. Year-to-year variability of air mass arrival to the region of the Laptev Sea from continents and from the Arctic in winter (for 850 hPa trajectories).

The mean frequencies of continental, arctic, and marine air masses arrival to the observation site, as well as the distributions (5 days later) of air outgoing to the continents, in the Arctic and the Pacific and Atlantic Oceans are given in Table 1. It follows from this Table: (1) about 50% of air masses comes to the observation site from the Arctic, (2) the frequency of air mass income from the Pacific and Atlantic Oceans does not exceed 5%, (3) air masses outgoing from the observation site in 20–40% of cases leaves the Arctic for five days (on the average, for 3.3 days).

In the cases, when air moves to the observation site from the continent over an industrial region emitting various anthropogenic pollutants into the atmosphere, these pollutants can be entrapped by air masses and transported to the Arctic. Such trajectories were considered separately for each source region depicted in Fig. 2, and statistical characteristics of the corresponding ensembles of trajectories were revealed.

Table 1. Spatial distributions of incoming and outgoing air masses, in %

Season	Incoming from			Outgoing to		
	continents	Arctic	oceans	continents	Arctic	oceans
Winter	56	42	2	30	60	10
Spring	45	51	4	29	67	4
Summer	46	49	5	38	62	0
Fall	49	50	1	20	80	0

The technique for estimating the contributions of individual industrial regions to anthropogenic pollution observed at the observation site in the Arctic was described in detail in Ref. 8. The contribution of a source region is proportional to its emission rate and the function called by us the function of transport efficiency (FTE). It characterizes the potential capability of a source to pollute the atmosphere of the observation site and depends only on the atmospheric processes along the path of air masses. The FTE depends on the probability and speed of air motion between the source and the observation site, which are estimated for an ensemble of trajectories, as well as on the conditions of vertical mixing and the rate of some pollutant removal from the atmosphere during the transportation.

Similarly to Ref. 8, the parameters needed to estimate FTE were chosen the same as for the Northern Land Archipelago (based on long-term meteorological data for the central part of the Russian Arctic). It was taken into account that the rate of dry deposition of a pollutant in the Arctic in winter and spring (over the surface covered by ice or snow) should be minimal,[9] and the rate of pollutant deposition with precipitation was calculated based on the long-term data on the intensity and the aggregate state of precipitations. Besides, we took into account the peculiarities of temperature inversions and cloudiness in the Arctic in different seasons.

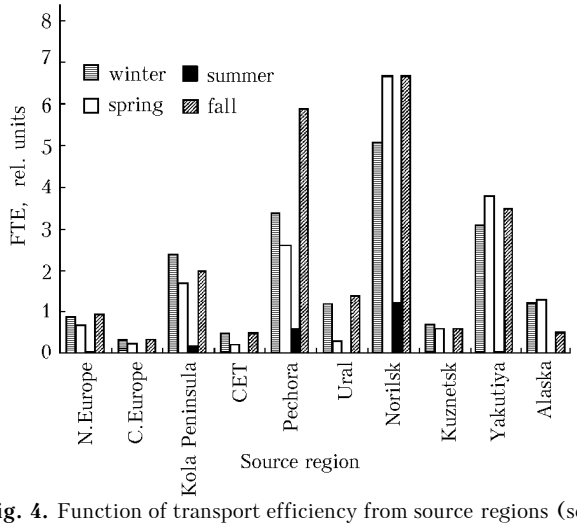

Fig. 4. Function of transport efficiency from source regions (see Fig. 2) to the observation site in different seasons, in rel. units.

Figure 4 depicts the nonzero FTE values for the considered observation site and thus demonstrates 10

industrial source regions polluting the atmosphere in the region of the Laptev Sea. It can be seen that in summer, as compared with winter and spring, the efficiency of pollutant transport to the Arctic decreases considerably, especially, from remote industrial regions. This is caused, on the one hand, by the northern displacement of the arctic front bounding the spread of the arctic air and, on the other hand, by a higher rate of pollutant deposition onto the surface in warm seasons, as compared with the cold ones.

The level of atmospheric pollution in the region of the Laptev Sea was estimated for six microelements (As, Ni, Pb, V, Zn, Cd), whose origin in the atmosphere is mostly related to the human activity. At long-range atmospheric transport, the effects of dry deposition and deposition with precipitation lead to more narrow size spectrum of aerosol particles,[9] thus decreasing and equalizing the deposition rates of different components (chemically inactive). Therefore, we assume that the deposition effects are the same for the considered chemical elements, and the deposition rate for them was taken the same as well. In calculation of winter and spring concentrations, we took into account that the pollution of air coming to the observation site from the Arctic in the cold period was roughly equal to that at the observation site. So the contribution of the arctic air can be thought proportional to the frequency of its income to the observation site (for details see Refs. 7 and 8). The contribution of the marine air to concentrations of anthropogenic components can be neglected.

Table 2. Most important sources of six anthropogenic microelements in the atmosphere over the Laptev Sea

Season	Element	Source	Contribution, %
Winter	As, Ni, Pb, V, Zn, Cd	Arctic	42
	As, Ni	Norilsk	23
	Pb, V	Ural	25
	Zn, Cd	Ural, Norilsk, Kuzbass	10–15
Spring	As, Ni, Pb, V, Zn, Cd	Arctic	50
	As, Ni, Pb	Norilsk	30
	V	Ural, Norilsk	12
	Zn	Kuzbass	24
	Cd	Norilsk, Kuzbass	15
Summer	As, Ni, Pb, V, Zn, Cd	Norilsk	60–80
Fall	As, Ni	Norilsk	50
	Pb, V	Ural	40–55
	Zn	Ural, Kuzbass	30–35
	Cd	Ural, Norilsk	25

Mean contributions of different source regions to atmospheric pollution in the region of the Laptev Sea are different for different elements and in different seasons. To estimate the contributions of the sources having nonzero FTE (see Fig. 4), we used the data on their emission rates with allowance for changes in the last two decades.[10] Industrial regions contributing most significantly to the atmospheric content of the considered six elements in the region of the Laptev Sea are listed in Table 2. Thus, the most significant source regions polluting the atmosphere of the Laptev Sea are the Norilsk region in summer, and Norilsk, Ural, and Kuzbass in other seasons.

Table 3 gives the mean (for each season) concentrations of the considered six chemical elements estimated as a sum of contributions from different sources and the contribution of the arctic air. It can be seen that the obtained values reasonably correspond to the available experimental data on the aerosol composition in the Arctic atmosphere. Field measurements show that in summer the atmospheric concentrations of the considered elements in the Arctic are roughly 10 to 50 times as low as in winter and spring.[4,11,13] In this connection, it can be concluded that our approach allows one to detect actual changes in the pollution level of the Arctic atmosphere in different seasons.

Table 3. Mean concentrations of six anthropogenic microelements in the atmosphere of the Laptev Sea in different seasons, ng·m^{-3}

Season, region	As	Ni	Pb	V	Zn	Cd
Winter[1)]	0.12	0.39	1.1	0.27	0.68	0.029
Spitsbergen[2)]	0.52	0.29	3.0	0.54	3.9	0.080
st. Alert, Canada[3)]	–	–	2	0.35	4	–
Spring[1)]	0.13	0.43	0.82	0.17	0.57	0.026
Northern Land[4)]	0.26–1.8	1.1–11	3.5	0.73–0.8	0.6–13	0.09
st. Alert, Canada[3)]	–	–	3	0.3	5	–
Summer[1)]	0.009	0.039	0.034	0.0084	0.016	0.0012
Spitsbergen[5)]	0.0168	<0.14	<1.0	0.041	0.59	<0.06
st. Alert, Canada[3)]	–	–	0.25	0.01	1	–
Fall[1)]	0.080	0.27	0.70	0.18	0.35	0.019
Laptev Sea[6)]	0.38	–	–	–	15.4	–
st. Alert, Canada[3)]	–	–	0.25	0.015	2	–

[1)] This work, calculation, means for 1986–1990.
[2)] Experiment, means for 1983, 1984 and 1986 (Ref. 11).
[3)] Experiment, means for 1981–1995 (Ref. 4).
[4)] Experiment, means for 1985, 1986, 1988 (Ref. 12).
[5)] Experiment, means for 1984, 1986 and 1987 (Ref. 13).
[6)] Experiment, means for 1995 (Ref. 1).

Knowing mean distributions of air masses in five days after their passage through the observation site (see Table 1) and the deposition rates, we can calculate, as was proposed in Ref. 7, the mean (for each month considered) spatial distributions of a pollutant after it leaves the observation site (Table 4). It can be seen that in winter after air passage over the Laptev Sea the pollutant fraction carried away by air masses for five days is almost three times as large as the fraction deposited onto the surface for this period in the Arctic region. In spring these fractions are roughly equal. Besides, in winter and spring about 50% of atmospheric pollutants brought to the region of the Laptev Sea live in the arctic atmosphere longer than five days, mixing there and then polluting the Arctic Ocean, its islands, and the coastal regions of the continents. In summer, to

the contrary, almost all pollutants deposit onto the surface of the Arctic Ocean during five days.

Table 4. Pollutant distribution in the Arctic in five days after departure from the observation site (%) at the corresponding deposition rates *K* in different seasons

Season	K, cm·s^{-1}	Leaves the Arctic	Deposits in the Arctic	Keeps in the atmosphere
Winter	0.05	37	11	52
Spring	0.10	27	24	49
Summer	0.98	3	96	1
Fall	0.42	4	89	7

Knowing the mean concentration of pollutants in the atmosphere (see Table 3) and the fraction deposited for five days onto the surface in the Arctic (see Table 4), we can estimate (similarly to Ref. 8) the mean flows of the chemical elements onto the surface in the vicinity of the observation site. The resulting estimates are given in Table 5. The annual estimates were obtained on the assumption that the winter values are valid for five months, summer ones – for three months, and spring and fall values – for two months of a year.[4] The area of the Laptev Sea was assumed equal to 662 000 km^2. It can be seen that the deposition from the atmosphere onto the surface of the Laptev Sea is maximal in spring (when the amount of precipitation increases at high atmospheric concentrations of pollutants) and in fall (when the concentrations of atmospheric pollutants begin to increase at still high precipitation amount).

It was traditionally thought that the amount of suspended matter (including the anthropogenic component) drained by rivers into the Arctic Ocean far exceeds the atmospheric contribution. The two lower rows of Table 5 show that this is not true for waters of the Laptev Sea basin. The amount of microelements in the suspended matter drained by the biggest rivers (Lena, Yana, and Khatanga) into the Laptev Sea was determined from estimates of mean annual flows[14] and experimental data[15] on the composition of suspended matter in these rivers. Besides, it was taken into account that only about 5% of the suspended matter drained by river water penetrates into the sea water beyond the marginal filter zone.[16] The role of such filter is played by a system of physical and chemical processes proceeding when mixing the fresh and salt waters at their confluence and leading to efficient deposition of suspended matter at the area up to tens of thousands of square kilometers near the mouth. For the Laptev Sea the total area of marginal filters of inflowing rivers does not exceed 5% of the sea area.

It can be seen from Table 5 that the flows of the considered metals coming to the central part of the Laptev Sea from the atmosphere and with the water of the inflowing rivers are comparable. It should be noted that the calculated mean contributions of lead and cadmium are far higher comparative to other elements. This may be related to the fact that in the arctic atmosphere just Pb and Cd can be attributed to anthropogenic origin to a greater degree than other four elements. Since the approach used in this work is developed to estimate the atmospheric concentrations of just anthropogenic components of aerosol or anthropogenic fractions in the concentration of constituents of mixed origin (for details see Ref. 8), the actual level of concentrations and flows of As, Ni, V, and Zn onto the surface may be higher (due to components of natural origin).

Thus, we have estimated the mean (for decade) levels of atmospheric concentrations of some heavy metals and their mean flows onto the surface in the Laptev Sea in different seasons. It has been shown that large industrial regions, such as Norilsk, Ural, and Kuzbass play the major role in atmospheric pollution in this region. The mean concentrations of six anthropogenic microelements in the atmosphere over the Laptev Sea in different seasons reasonably correspond to similar characteristics determined in other arctic regions. The monthly mean flows of anthropogenic components of the atmosphere onto the Laptev Sea surface are maximal in spring and fall and minimal in summer. The yearly average contribution of atmospheric aerosols to the content of anthropogenic pollutants in water of the central part of the Laptev Sea (and, consequently, in bottom deposits) may achieve one third of the contribution of inflowing rivers.

Table 5. Mean flows of six anthropogenic microelements from the atmosphere to the surface of the Laptev Sea in different seasons and for a year

Season	As	Ni	Pb	V	Zn	Cd	Parameter
Winter	0.10	0.34	0.95	0.23	0.59	0.025	Flow per unit area, g·km^{-2}·month^{-1}
Spring	0.21	0.68	1.3	0.27	0.91	0.041	
Summer	0.04	0.18	0.16	0.04	0.07	0.006	
Fall	0.27	0.90	2.3	0.60	1.3	0.063	
Year	1.6	5.4	12.4	3.0	8.9	0.35	g·km^{-2}·year^{-1}
Winter	70	220	630	150	400	16	Flow to the Laptev Sea as a whole, kg·month^{-1}
Spring	140	450	860	180	600	27	
Summer	30	120	100	25	50	4	
Fall	180	600	1500	400	860	42	
Year	1.1	3.5	8.2	2.0	5.1	0.23	t·year^{-1}
	13.5	60	26	135	160	0.65	River run-off*, t·year^{-1}
	7.5	5.5	24	1.5	3.5	26	Aerosol contribution, %

* As estimated from the data on flows and composition of river suspended matter from Refs. 14 and 15.

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