

Arctic aerosols. Results of ten-year investigations

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The results of investigations of above-water aerosols over seas of the Russian Arctic, Central Arctic, and in the North Pole region are presented. The investigations were conducted during 10 research missions for the period from 1991 to 1998. The mineral and chemical composition of aerosols, their particle size distribution, transport ways of the aeolian material, and the degree of anthropogenic pollution of the arctic atmosphere were studied. A particular attention was paid to estimation of horizontal and vertical flows of various components. The studies were conducted simultaneously by several methods: netting particles with nylon nets of large (10 m²) area, air filtering through the Whatman-41 and AFA-KhA filters, and counting particles with photoelectric counters.

Introduction

Arctic aerosols are poorly known. Meanwhile, as compared to other climatic zones, the Arctic is characterized by specific conditions determining the amount, properties, and composition of the aerosol material. First of all, this is a high-latitude position of the Arctic – in the region of global divergence in the cells of vertical circulation. The stratospheric aerosol material (maximum content at the altitude about 20 km) is mixed there with the tropospheric one and drops to the cloudy ground layers, wherefrom this material deposits. This model is supported by results of studying the radioactive fallout after H-testing¹ (Fig. 1). The meridional circulation is more intense in winter.²

The conditions of the aerosol carry-over from the land and sea surface differ markedly. In winter the land surface in the Arctic is ice-bound and covered with snow throughout many thousands kilometers, and the sea surface, which usually delivers a great deal of sea salt to the atmosphere, is also covered with ice. The influence of local and regional sources of the aerosol material (both marine and continental) is thus excluded for a long time.

Other unusual conditions are connected with authigenic aerosols, which are generated in the upper atmosphere affected by cosmic rays under the conditions of very strong electromagnetic fields (group of cosmogenic isotopes, etc.). Probably, just here – in the Arctic and Antarctic – the contribution of the meteoritic matter, entering the Earth's atmosphere, can be best estimated, as well as the influence of the global background of the planet.

There is some evidence that the aeolian material falling out of the atmosphere is the main source of biogenic elements in the Arctic, as well as iron and some metals important for evolution of life. Of great interest is also the study of various anthropogenic

pollutants coming to the Arctic from far industrial centers located at the distance more than 1000 and even 10000 km away. Finally, recent satellite investigations of the land and ocean surface, in particular, determination of the chlorophyll content in the surface water, require correction for the atmospheric layer, what is impossible without comprehensive study of not only quantity, but also quality (composition) of the aerosol material.

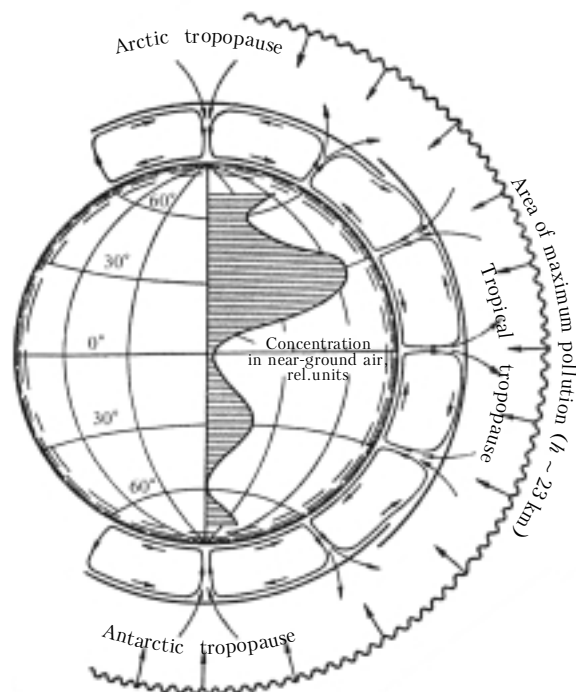


Fig. 1. Meridional circulation of air and distribution of radioactive fallout (shaded) at the stratospheric type of transport of sedimentary material.¹

Besides, the arctic aerosols are of particular importance, because most rivers in the Arctic (50–80%

of Siberian rivers) are supplied by snow. At spring melting, water runs down the frozen land surface, so the effect of the soil itself and the aeoliation crust on the composition of thawing water shows itself only at the end of the spring flood. Thus, the composition of the river water here is largely determined by the composition of snow – the principal agent of aerosol washing out from the atmosphere in the winter season, that is, 200–300 days a year.

Another peculiarity is connected with the ice cover of the Arctic. Every year the depth of the snow cover here is about 30–40 cm. In summer the snow on the ice surface melts, forming fresh water pools, and the aerosol matter from the thawing water is concentrated at their bottom. Some water pours down through cracks and current holes, but the temperature of the sea water in polynias and under drifting ice is below zero (from –1.7 to –1.8°C). The fresh water pouring down from ice freezes again and thus increases the bottom part of the ice (the aerosol is transformed into cryosol). Thus, the aerosol material turns out to be closely connected with the drifting ice, gradually penetrates into their depth, and comes to the sea water only after melting – this exposition often lasts up to 10–15 years. The arctic ice cover is an accumulator of the aerosol matter and its additional carrier. The aerosol material is concentrated at the places of ice melting (unloading) first in the water suspension and then in bottom sediments.

The aerosol influence on the Arctic biota is also significant. Biogenic elements, needed for development of phytoplankton, fall with aerosols on the land and sea ice surface. It should be noted that in the Arctic the river water delivers much less biogens than in other climatic zones, what is connected with the absence of *Azotobacter* in the Arctic soil (because of the low temperature), as well as with poor development of chemical aeoliation. Just aerosols provide for phytoplankton florescence in the fresh water pools on ice and plankton vigorous development on the edge of ice.

The aerosol has also a significant influence on the process of sedimentation in the Arctic Ocean and its seas, because the arctic rivers carry 10–20 times less amount of suspended matter comparative to the globally mean value (20–30 mg/l at the globally mean value about 500 mg/l) (Refs. 3 and 4). Thus, the Arctic aerosols play an important role in formation of the environmental conditions, in particular, in the climate formation.

By the beginning of our investigations, one interesting, unusual for other zones, regularity has been already found in the quantitative distribution of aerosols. Maximum amounts of aerosol were measured not in summer, when the land surface is free of snow and can deliver mineral and plant material, but in winter, when the land and islands are covered with snow. The estimated excess reached 5–10 times.

The most part of the Arctic territory is occupied by seas, but the data on peculiarities of aerosol distribution above seas in high latitudes are almost

absent – most observations have been conducted at polar stations situated on the land or large islands.

Formerly, aerosols have been studied in Siberia, on Wrangel Island, the Northern Land, the Franz Josef Land, Spitsbergen, Greenland, in Alaska, and in the polar areas of Canada.^{5–21} The systematic studies of quantitative distribution, composition, and other characteristics of the aerosol material were started by us in 1991 during marine missions in the seas of the Russian Arctic and are still in progress. Nevertheless, very few results have been published by now.^{22–30}

Previously, the Arctic aerosol was mostly studied as a probable source of pollution of the polar ecosystem.^{6,7,10,18,31–33} Our approach to the study of the aerosol differs – most attention is paid to the role of the aeolian material in formation of the environment and climate of the Arctic and its influence on the biota, ice, process of sedimentation, geochemistry, and lithology. This paper for the first time presents some elements of our new approach and, in particular, shows the former ideas on a modest role of the aerosol material in formation of the Arctic environment^{34–36} to be wrong. The delivery of the aerosol material on the surface of drifting ice and in bottom sediments, as well as in the ice-bound river basins (in the form of snow) prove to be significant and even principal for many elements. The actual scales of this process are being brought to light only now.

The Arctic aerosol has been studied within the confines of the unified global program^{37–40,etc.} and by the common method, what allows its comparison with other climatic zones. Such a comparison was started at the Institute of Oceanology about 40 years ago.

1. Materials and methods

The Arctic aerosols in the above-water atmospheric layer are studied for ten years (1990–1999) by the specialists from the Laboratory of Physical-Geological Research of the Institute of Oceanology of the Russian Academy of Sciences. Below we present the results obtained largely for the Western and Central Arctic in comparison with the literature data for other regions.

At least four important methodical aspects of our works should be mentioned:

First, for comprehensive study of the amount and composition of aerosols we used a system of complementary and sometimes even overlapping methods, namely,

- the methods of air filtering through specialized filters,

- the netting of large amounts of a suspended matter into nylon nets using static electric charges,

- the determination of the aerosol particle size spectrum by photoelectric counters of various types,

- the determination of vertical aerosol flows by isotopic methods, as well as of trajectories of air and aerosol masses by the back plotting method using a set of meteorological maps and the data of point observations.

Second, in our studies we widely employed a system of sedimentation methods: collection and study of aerosol material deposited with snow, located on the surface and in the depth of pack ice, land glaciers, bottom sediments of seas and oceans (in some cases, lake sediments as well).

These methods allow the data to be collected not only at the moment of vessel observations, but for long periods of time: months, seasons, years, decades, centuries, and millennia. Such long observation series in the Arctic are usually absent or are possible only in rare cases. This is especially important because the studies can be conducted not at stationary stations, but on board moving vessel, often in route sections of more than 100 km long. Thus, all types of natural recorders are invoked to aerosol studies, and they acquire a complex character. The studies allow the role of the aeolian material in formation of the Arctic environment to be understood to a greater extent. Since the time coordinate is also invoked, then for the first time the possibility of the 4D analysis of the Arctic aerosol system is opened up.

The third peculiarity is that the measurements, along with the data of the ship-borne meteorological station, were compared with collections of the synoptic maps of the region given by the Gidrometeosluzhba (Russian Hydrology and Meteorology Service). This allows reconstructing the air mass paths, revealing places of probable entrainment of aerosols and the places of their unloading above the Arctic water area, that is, the dynamic analysis of the aerosol system.

The fourth peculiarity is that, in contrast to traditional studies, we paid our attention to quantitative examination of aerosol flows and their different components, including pollutants. For this purpose, in addition to direct methods (examination of snow fallen for a certain time), the analysis by precipitation-collecting systems as well as sedimentation and isotopic (^{210}Pb , ^7Be , ^{222}Rn , and others) methods were used. The combination of the methods of studying vertical and horizontal aerosol flows is especially important in the complex analysis of air, ice, water, and sediment reservoirs in their interaction, what allows monitoring the atmosphere on a new basis.

Air was filtered through AFA-KhA-20 acetate-cellulose filters with the working surface of 20 cm² (Ref. 41) and Whatman-41 glass fiber filters, which trap also submicron particles, including particles of sea salt.

The amount of the aerosol material collected by the filters was insufficient for studying the mineral composition, and, in addition, the material itself was clogged with sea salt. Therefore, filtering was accompanied by netting the aerosol with nylon nets.^{38,42} This method allows collection of large amounts of aerosols. The nets were set on the bows of the vessel at the altitude of 4 to 9 m above the deck; the total net area was about 10 m². The counter air flow induced the electrostatic charge, which attracted

particles. However, as the special-purpose investigation has shown, not all finest particles are netted. After the exposition (usually one day long) the nets were cleaned by flushing with distilled water, the charge was removed, and the particles were separated from the water by filtering through a nuclear filter with the pore size of 0.45 μm . These samples were not contaminated with sea salt. Sites of netting are mapped in Fig. 2. A total of 68 samples was collected in such a way. In some cases, sampling was hampered by rain and snow as well as storm conditions. In parallel, 53 samples were collected by filtering, and these samples were mostly used for studying the chemical composition.

The aerosol particle size distribution was examined by analyzing the filtered samples with a scanning electronic microscope and using PC-218 photoelectric counter (Royco, USA; ranges of 0.5–1, 1–2, 2–3, 3–5, 5–10 μm). In our research missions, the size distribution was determined in 320 cases (Fig. 3).

The third independent method was used for parallel analysis of the size spectrum of the samples collected by nets. The suspension was separated from the nuclear filters in an ultrasonic cell⁴³ and then analyzed in the range of 0.5–100 μm with the use of the CIS-1 laser electrooptical particle counter (Galai, Israel) at the A. Vegener Institute of Polar and Marine Research (Germany).

The main method for studying the chemical composition of aerosols was the instrumental neutron-activation analysis (INAA).^{44,45} For determination of some elements, the method of atomic absorption with the Perkin–Elmer-3030 device having a graphite cell was used (at the Institute of Marine Biochemistry, France). A total of 121 samples was analyzed by the INAA, and 10 samples were analyzed by the method of atomic absorption. In many samples, some individual characteristic particles were examined with the JXA-733 microprobe (Japan) at the Laboratory headed by Professor Van Grieken.²⁷ The element composition of 400 particles with the sizes from 0.4 to 15 μm was studied in each sample in an unattended mode.⁴⁶ A total of 10 samples was examined in such a way.

The aerosol composition was also studied with a scanning electron microscope, and the mineralogical analysis of the netted samples was made by the method of immersion as well as X-ray methods.^{24,47}

The possible sources of aerosols and the trajectories of aerosol income to the Arctic were revealed by analyzing the collection of meteorological maps for the last ten years^{48,49} as well as by calculating direct and back trajectories of air motion to the Russian Arctic for April – July, 1986–1995, based on the data of the Russian Hydrology and Meteorology Center.⁵⁰ This analysis is now completed for three Arctic regions (the Franz Josef Land, Wrangel Island, and the Northern Land).

The sedimentation methods of aerosol study (snow – ice – water – bottom sediments) are described in a number of papers.^{51–53}

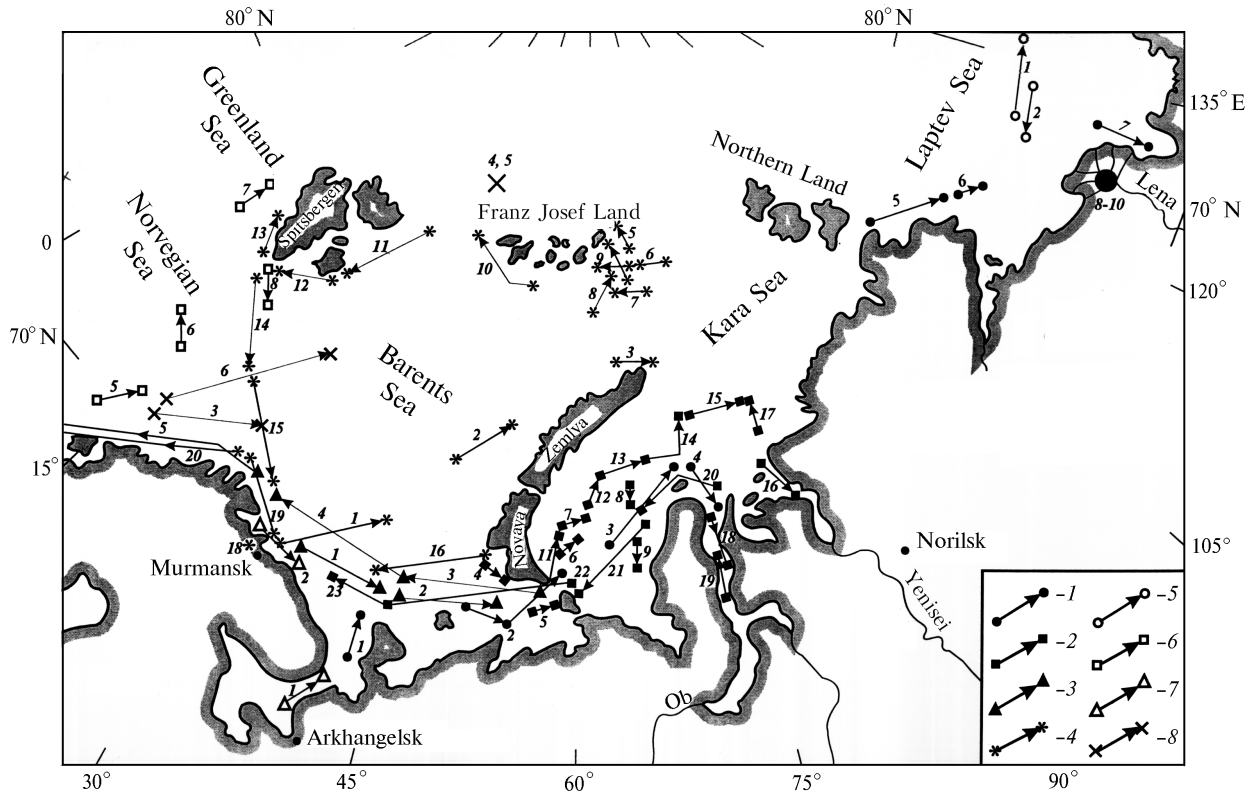


Fig. 2. Netting samples of Arctic aerosols: SPASIBA-91 (1), the 49th trip of research vessel (RV) *Dmitrii Mendeleev* (2), the 31th trip of RV *Akademik Mstislav Keldysh* (3), the 9th trip of RV *Professor Logachev* (4), ARK-XI/1 trip of RV *Polarstern* (5), the 15th trip of RV *Professor Logachev* (6), the 11th trip of RV *Akademik Sergei Vavilov* (7), the 14th trip of RV *Akademik Fyodorov* (8).

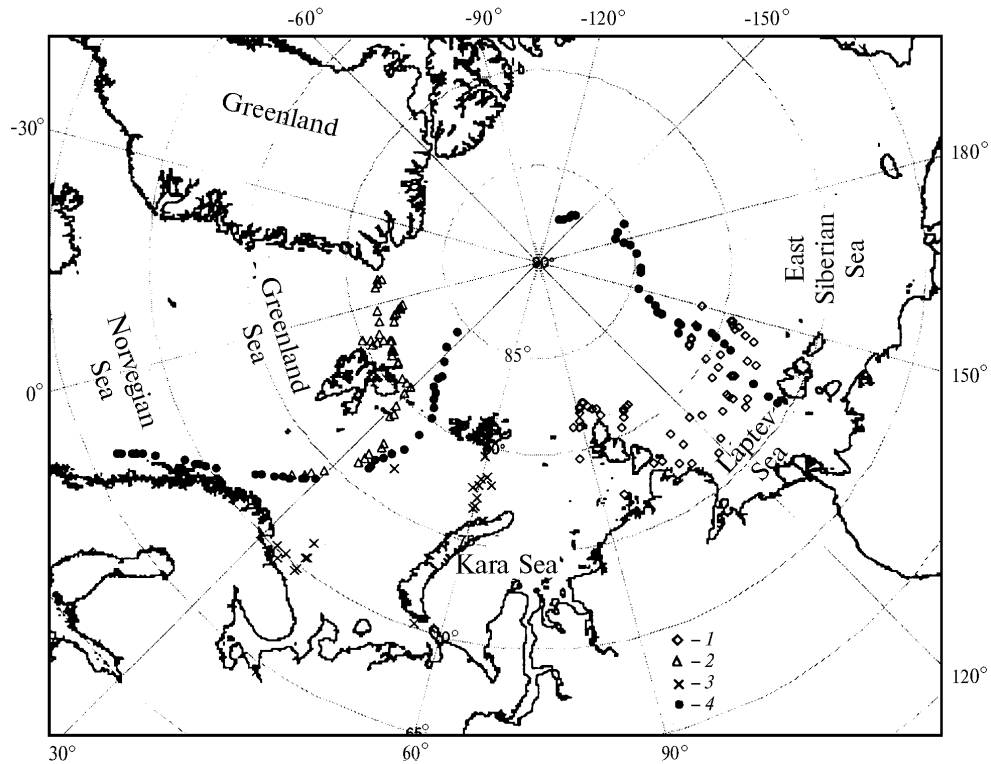


Fig. 3. Sites at which the particle size distribution of Arctic aerosols was determined: ARK-XI/1 trip of RV *Polarstern* (July-September 1995) (1), ARK-XIII/2 trip of RV *Polarstern* (July-August 1997) (2), the 11th trip of RV *Akademik Sergei Vavilov* (August-October 1997) (3), ARK-XIV/1a trip of RV *Polarstern* (July 1998) (4).

2. Results and discussion

2.1. Quantitative distribution of aerosols

The content of the insoluble (salt-less) part of the aeolian suspension depends on the climatic zone, closeness of a large land, wind speed and direction, air humidity, and atmospheric precipitation.^{38,40,54–57} The aerosol distribution in the Russian Arctic is described in a number of our papers.^{22,25,29,30,58}

The highest content of water-insoluble particles ($1.38 \mu\text{g}/\text{m}^3$) was observed from 19 to 20 of August, 1996, in the Greenland Sea at the north-westerly wind flow from the Greenland. The analysis of the sample with an electron microscope and the study of the chemical composition of this sample have shown that the terrigenous material dominates in it. This material was blown out of the soil and open rocks of the coastal areas of the Greenland by strong winds (fans). Very strong (katabatic) winds at air masses running down from the ice dome of the Greenland are characteristic of this region,⁵⁹ which deliver a large amount of relatively large particles ($> 2 \mu\text{m}$) of soil origin into the coastal atmosphere.⁶⁰ The lowest content of the aeolian suspension ($0.02\text{--}0.06 \mu\text{g}/\text{m}^3$) was observed in the case of sampling away from the coast after rain, what is a characteristic of aerosol washing-out by atmospheric precipitation.⁶¹

The mean content of water-insoluble aerosol particles in the ground atmospheric layer of the Russian Arctic (averaged over 55 cases) is $0.23 \mu\text{g}/\text{m}^3$ at the standard deviation of $0.19 \mu\text{g}/\text{m}^3$. This is higher than the literature data on concentrations in open ocean

areas, especially, in humid zones, but much lower than the suspension content in arid zones (Table 1).

The aerosol content in the Global Ocean depends on a climatic zone and the distance from the land (circumcontinental zoning) and ranges from 0.01 to $0.1 \mu\text{g}/\text{m}^3$ in open areas of the northern humid zone of the Atlantic Ocean, from 0.02 to $2.34 \mu\text{g}/\text{m}^3$ in the western part of the equatorial humid zone of the Pacific Ocean near islands, and from 10 to $50 \mu\text{g}/\text{m}^3$ in the coastal regions of the northern arid zone of the Atlantic Ocean.^{40,57} The content of the aeolian suspension in the Arctic is characterized by wide spatial and temporal variability.

In the regions of our investigations in the seas of the Russian Arctic, the circumcontinental zoning was not pronounced, because the routes of the vessels passed mostly near the land (Eurasia or large islands) (see Fig. 2). However, the high content of insoluble aerosols was observed under conditions of dry weather in the regions closest to the land, especially, at the wind blowing from the land. For example, in August of 1991 during the SPASIBA-91 mission the highest concentrations of the aeolian suspension ($0.40\text{--}0.46 \mu\text{g}/\text{m}^3$) were measured in the vicinity of the continent at a strong wind ($12\text{--}15.7 \text{ m/s}$) blowing from the land. The aerosol size distribution spectrum varies fast and depends on the combination of many local and regional factors.

In summer the aerosol content is strongly affected by sea salt, whose percentage achieves $70\text{--}80\%$ of suspension on the filters.^{56,62} The salt content increases with increasing the wind speed and decreasing the air humidity.

Table 1. Content of insoluble fraction of aerosols in the above-water atmospheric layer over seas of the Russian Arctic in comparison with other regions of the Global Ocean

Region	Number of samples, n	Content, $\mu\text{g}/\text{m}^3$				Source
		min	max	mean	standard deviation	
Russian Arctic	55	0.02	0.97	0.23	0.19	This work
Northern humid zone of the Atlantic ocean, open areas	–	0.01	0.1	–	–	Ref. 40
The same, coastal regions	–	–	20	–	–	Ref. 40
Northern arid zone of the Atlantic Ocean, open areas	–	–	–	7.7	–	Ref. 40
The same, coastal regions	3	5.8	11	–	–	Ref. 42
Northern arid zone of the Indian Ocean, coastal regions	4	2	4.6	–	–	Ref. 39
Equatorial zone of the Atlantic Ocean, open areas	3	0.92	4.4	–	–	Ref. 98
Equatorial zone of the Indian Ocean	–	–	–	0.61	–	Ref. 40
Equatorial zone of the Pacific Ocean, western part	7	0.01	0.25	–	–	Ref. 39
Equatorial zone of the Pacific Ocean, western part	3	0.01	0.25	–	–	Ref. 98
Southern arid zone of the Atlantic Ocean, coastal region near Namibia	22	0.02	2.34	0.46	0.55	Ref. 100
Southern arid zone of the Atlantic Ocean, coastal region near Namibia	2	3.58	5.68	4.63	1.48	Ref. 99

2.2. Aerosol particle size distribution

From the measurements at 63 sites, the content of particles larger than $0.5 \mu\text{m}$ over the Laptev Sea during the ARK-XI/1 mission of the ice-breaker *Polarstern* (July–September, 1995) varied from 0.09 to 21.4 cm^{-3} . On the average, it was 7.84 cm^{-3} at the standard deviation of 6.47 cm^{-3} (Ref. 63). These values are close to the number density of particles of the same size in the Norwegian Sea, on Wrangel Island, on the Franz Josef Land, and in the central region of Russia (Obninsk)^{23,64–67} and far lower than those in the arid regions of Tadjhikistan (from 20 to 100 cm^{-3}) (Ref. 68).

On the whole, the particle size distribution in the surface atmospheric layer above the Laptev Sea and other Arctic regions differs insignificantly from that above Obninsk and other land areas (see Fig. 4).

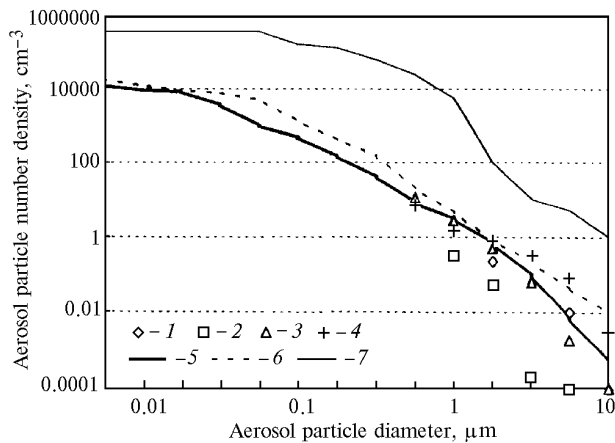


Fig. 4. Cumulative curve of the aerosol particle size distribution in the surface atmospheric layer above the Laptev Sea from July, 20 to September, 10 of 1995 (data of ARK-XI/1 trip of the German research vessel *Polarstern*) and other regions²³: Laptev Sea: all days (1), fine days (2), days with strong wind (3), days with fog (4); Franz Josef Land, spring 1994 (5); Obninsk, Kaluga Region, May 1994 (6); dried-up bottom of the Aral Sea, Kazakhstan, dust storm of May 30 of 1992 (7).

In the general case, the content of fine particles (from 0.5 to $1.0 \mu\text{m}$) is much higher than that of coarse particles (Fig. 4). In the regions where open (free of ice) water occupies more than 30% of surface, the content of particles larger than $0.5 \mu\text{m}$ increases markedly with increasing the wind speed (Fig. 5).^{23,28,63} This indicates the income of sea salt particles to the atmosphere from the thin surface layer of the sea water (what is also confirmed by the element composition of corresponding aerosol samples). The increasing content of particles in the surface atmospheric layer at the increasing wind speed was earlier noticed in other ocean areas.^{65,69,70} In the northeastern Laptev Sea at the wind speed higher than 8 m/s the content of particles $> 0.5 \mu\text{m}$ varied from 1.05 to 11.23 cm^{-3} , making on the average 6.52 cm^{-3} (from the data of 30 measurements at 10 sites), while at the wind speed of 3.3 m/s it was only 0.09 cm^{-3} (Ref. 63).

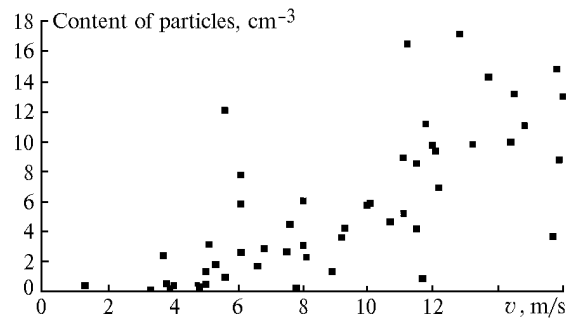


Fig. 5. Influence of wind speed on the content of particles $> 0.5 \mu\text{m}$ in the surface atmospheric layer over the Laptev Sea in July–September of 1995 (ARK-XI/1 trip of *Polarstern*) (Ref. 63).

In generation of fog, the total content of particles larger than $0.5 \mu\text{m}$ remains at the standard level, whereas the number of particles with the size from 2 to $5 \mu\text{m}$ increases sharply (Fig. 6) due to formation of large fog droplets and condensation of smaller particles. This phenomenon was earlier studied under both field and laboratory conditions^{71–73}; it is very significant for washing out particles from the atmosphere (suspended, colloid, and dissolved substances).

In the ice-covered northwestern part of the Barents Sea near the underwater Plateau Ermak and in the Fram Strait, the content of particles with the size from 1 to $2 \mu\text{m}$ is higher than in ice-free sea, especially, at the temperature about -3°C (Ref. 74). This is likely the result of ice microcrystals growth in the near-water atmospheric layer at lowering the temperature.⁷⁵ The tentative processing of materials obtained in the ARK-XIV/1 trip of the *Polarstern* in the Central Arctic in July, 1998, has revealed the same regularities in the aerosol particle size distribution.⁷⁶

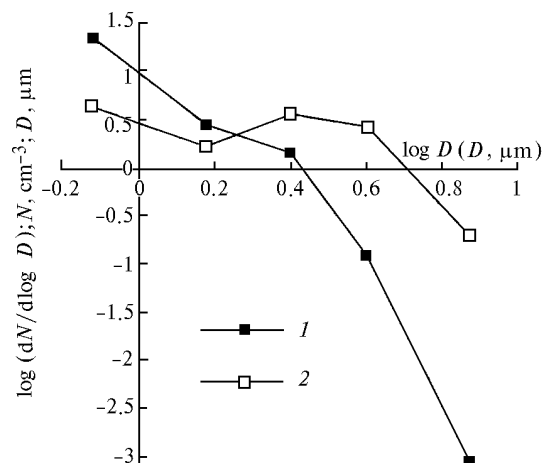


Fig. 6. Influence of weather (humidity) on the size spectrum of aerosols above the Laptev Sea in July–September of 1995 (ARK-XI/1 trip of the *Polarstern*): fine days (1) and foggy days (2).⁶³

The size spectrum of insoluble particles in 30 aerosol samples obtained by netting in the Arctic Seas was examined at a coastal laboratory with the Galai-CIS-1 laser counter.²⁹ The mean size of particles varied

insignificantly from sample to sample (from 1.24 to 1.69 μm). Large particles (from 5 to 25 μm) made the maximum contribution to the sample volume (and, consequently, mass).

Generalization of the results on the aerosol particle size spectrum made with particle counters both *in situ* and in the laboratory as well as the qualitative estimation of the particle sizes with a scanning electron microscope have shown that submicron salt particles dominate in air, but sediments are mostly (in mass) formed by water-insoluble particles with the sizes from 5 to 25, rarely to 50 μm .

2.3. Composition of aerosol matter from data of analysis with an electron microscope

According to the data of an electron microscope,^{22,25,29,58} coarse ($> 1 \mu\text{m}$) non-salt particles of Arctic aerosols sampled by netting consist largely of mineral and organic matter (plant fibers, pollen, diatom alga, etc.).

Land soil is the main source of mineral particles. In winter the main contribution is made by far transfer.^{77,78} Plant fibers up to several hundreds micrometers long and pollen of land plants are transported by wind to the distances of hundreds kilometers. Plant fibers are wind-blown out of the surface of the Arctic soil rich in plant residues.^{79,80} Pollen and spores detected in the water suspension and bottom sediments of Arctic seas^{81,82} come on the sea surface mostly due to the aeolian transport.

Some samples contain diatoms. Marine diatoms enter the near-water atmospheric layer due to the wind blowing-out from the sea water surface, whereas fresh-water diatoms come due to the far transport of dust from the surface of dried-up tundra lakes.^{83,84} Earlier the diatoms were found in aerosol samples from other regions of the Global Ocean.⁸⁵ Fresh-water and sea diatoms can come to the ice dome of the Antarctica in the vicinity of the South Pole and the ice dome of the Greenland only by the aeolian way.^{86,87}

Porous fly ash particles from 5 to 50 μm in size and smooth spheres from 0.5 to 10 μm in diameter are also characteristic of the Arctic aerosols. These particles were found in small amounts in most netted samples. Porous carbon particles mostly enter the atmosphere through emissions of metallurgical and mining plants, as well as power stations. Another source is forest fires.^{88,89} During all the research missions, when approaching the Kola Peninsula and Norilsk, the content of porous ash particles in aerosols increased. These particles are also accumulated in the Arctic soil, glaciers, and lake bottom sediments; they are indicators of the aerosol transfer of pollutants.⁹⁰ The studies of the surface layer of bottom sediments in the lakes located far from industrial centers (Spitsbergen, the Shetland Islands, northern regions of Norway, Sweden, and Finland) have shown that the porous carbon particles are present in all the lakes under study and their content is at the background level.^{91,92} The content of the porous carbon particles increases sharply in the surface layer of the bottom sediments in

the lakes of the Kola Peninsula located in the vicinity of mining and metallurgical plants.⁹² Their relatively high content was found in the surface layer of bottom sediments in lakes of other industrial regions, in particular, in sediments of the Great Lakes in the USA.⁹³

Smooth spheres of 0.5–10 μm in diameter are formed at high-temperature processes, and they are often referred as combustion spheres.⁹⁴ They are emitted into the atmosphere not only by metallurgical plants, but also by some other anthropogenic sources, for example, at the combustion of oil and hard coal, what is confirmed by their chemical composition.⁹⁵ They can be transported by air masses to long distances (they were found in Arctic aerosols^{94,96}).

The highest content of the spheres (more than 50% of particles larger than 1 μm) was found by us in the sample 16 taken from the southern Kara Sea in September, 1993, (the 49th trip of *Dmitrii Mendeleev*) under dominant income of air masses from the Norilsk region (according to the maps of baric situations), see Fig. 7. This sample was also heavily polluted by other aerosol particles formed in the smog emitted by the Norilsk Copper-Nickel Plant (with anomalous chemical composition of both the combustion spheres and the sample as a whole).

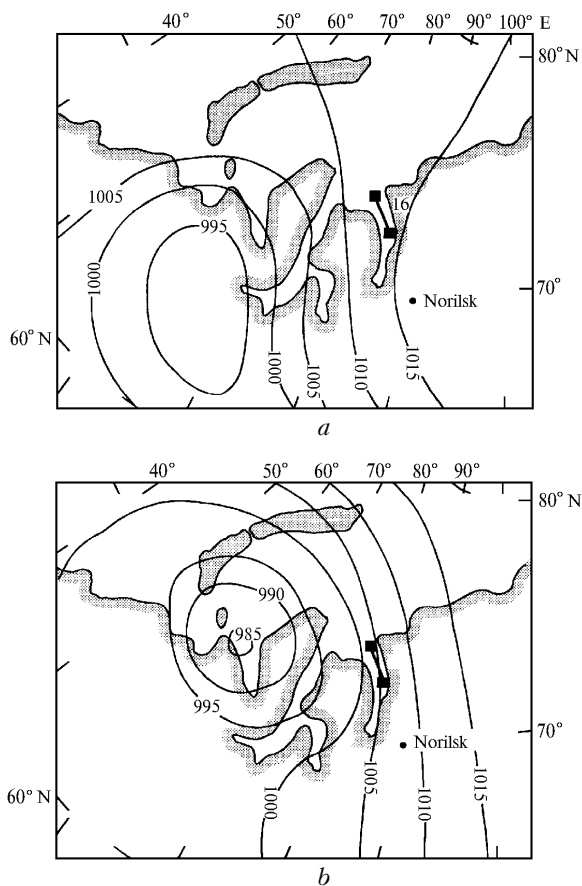


Fig. 7. Baric situations over the Kara Sea and adjacent regions in the beginning and end of netting in the 16th and 49th trips of RV *Dmitrii Mendeleev*: (a) 06:00 UTC, September 17, 1993, and (b) 06:00 UTC, September 18, 1993.

Thus, the study of the morphology of coarse ($> 1 \mu\text{m}$) insoluble particles and biogenic residues with an electron microscope indicates that the main source of the aeolian material over the Arctic seas in July–September is Eurasia, although in winter the contribution of far and very far ($> 10\,000 \text{ km}$) sources increases significantly.

2.4. Mineral composition

The mineral composition of aerosols is one of the important indicators of sources of the aeolian material^{40,47,97,98}; it also affects the optics and chemical composition of aerosol. The percentage of minerals collected by nylon nets was calculated for the crystal phase. The material under study consisted largely of particles from 1 to $10 \mu\text{m}$ in size.^{24,47} The characteristic of the aerosol mineral composition in our samples was its high spatial and temporal variability.

Quartz is the main mineral of the Arctic aerosols in the insoluble phase. Its content averaged 61.5% in mid-August–early September of 1991 (of the crystal phase), 33.2% in August–September of 1993, and 31.1% in August–October of 1994. On the average (the analysis of 47 samples collected in August–October in three missions), the Arctic aerosols contain 36.8% of quartz (at the standard deviation of 16.1%) (Table 2). The highest quartz content was measured in the samples collected near the continent, and the lowest one was in the open sea far away from the coastal areas.

The content of feldspar averaged over all the samples was 9.7% (at the standard deviation of 5.2%) (see Table 2). One of the important characteristics of the aerosol mineral composition is the quartz/feldspar

ratio (Q/F). The value of Q/F depends on the intensity of the aeoliation processes proceeding with the prevailing destruction of feldspar and conservation of quartz.⁴⁰ In shale – the main sedimentary rock of continents – the value of Q/F is close to 2 (Ref. 102). The increase of this ratio is indicative of more intense destruction (aeoliation) of feldspar, which is especially significant in tropics. For example, in the northern part of the Indian Ocean (arid zone) the quartz/feldspar ratio most often varies from 2 to 3 (Ref. 47), and in the east of the equatorial zone of the Pacific Ocean in April–June of 1990 it was, on the average, 3.7 (Ref. 100). This ratio is even higher (6.8) in the eastern part of the equatorial zone of the Atlantic Ocean near Africa.⁹⁹ In the Russian Arctic the surprisingly high values of the quartz/feldspar were measured. The Q/F ratio averaged over 47 samples was equal to 3.9, which indicated that the mineral suspension in the Arctic seas originated from regions with intense aeoliation of the feldspar from soils, that is, remote sources.

Among clay minerals, illite, chlorite, and kaolinite are the most abundant in the Arctic aerosols (see Table 2). The illite is the main clay mineral in the Arctic aerosols, megaprovince of the continental core, suspension of most rivers (except for rivers of the equatorial zone, in which the concentrations of kaolinite and montmorillonite are high).¹⁰³ If the sum of the clay minerals is taken as 100%, then, according to our data, the illite in the netted samples of aerosols of the Russian Arctic was, on the average, 50.9% in August–September of 1991, 41.5% in August–October of 1993, 45.5% in August–October of 1994; for all the Arctic trips the value averaged over 47 samples was 42.9% of the sum of clay minerals (at the standard deviation of 10.1%) (see Table 2).

Table 2. Mean mineral composition of aeolian suspension in the surface atmospheric layer over seas of the Russian Arctic in comparison with other regions of the Global Ocean

Missions, studied regions	Number of samples, <i>n</i>	Content of minerals in crystal phase, in %										Quartz/feldspar	Content of minerals, in % (of sum of clay minerals)						Source
		Quartz		Feldspar		Illite		Chlorite + kaolinite		Montmorillonite			Illite		Chlorite + kaolinite		Montmorillonite		
		av.	rms	av.	rms	av.	rms	av.	rms	av.	rms		av.	rms	av.	rms	av.	rms	
SPASIBA-91 ¹	9	61.6	8.7	11.8	3.8	13.6	3.0	13.1	5.4	–	–	5.2	52.3	12.4	47.7	12.4	–	–	Ref. 27
DM-49 ²	19	33.3	10.7	11.9	4.9	22.9	6.8	30.9	8.9	1.3	0.5	2.8	42.5	7.6	56.8	7.4	0.7	1.2	This work
Pl-9 ³	19	31.1	13.0	6.4	4.3	29.5	9.7	29.0	7.2	6.2	2.8	4.9	46.0	10.1	46.6	7.9	7.4	6.8	Ref. 26
Russian Arctic ⁴	47	37.8	16.1	9.7	5.2	23.8	9.6	26.8	10.2	4.7	3.3	3.9	45.8	10.1	50.9	9.8	3.3	5.6	This work
Equatorial Atlantic ⁵	4	37.3	3.8	5.5	4.5	22.5	5.4	31.3	2.9	3.4	1.2	6.8	39.3		54.7		6.0		Ref. 99
Equatorial part of the Pacific Ocean ⁶	13	51.9	13.1	13.9	4.2	6.9	2.3	27.3	11.2			3.7	20.2		79.8				Ref. 100
Tyrrhenian Sea ⁷													67		33				Ref. 101
NE Atlantic ⁸													45		41		14		Ref. 97

¹SPASIBA-91 is the international mission on studying the Arctic and Siberian water area and Laptev Sea in August–September 1991.

²The 49th trip of the RV *Dmitrii Mendeleev*, Kara Sea, August–October 1993.

³The 9th trip of the RV *Professor Logachev*, Barents Sea, St. Anna Channel, August–October 1994.

⁴The values averaged over the results of the three above-mentioned missions.

⁵The equatorial part of the eastern Atlantic, September–October 1990.

⁶The western Pacific Ocean, equatorial humid zone, April–June 1990.

⁷Tyrrhenian Sea at air masses coming from Europe.

⁸The northeastern Atlantic between 05°N and 35°N, arid zone.

For comparison, the illite content in aerosols of the equatorial humid zone (tropics of the western Pacific Ocean in April–June of 1990) was only 20.2% of clay minerals,¹⁰⁰ whereas in the temperate humid zone of the Northern Hemisphere (Tyrrhenian Sea, at air mass coming from Europe) it was 67% (Ref. 101).

The content of two other clay minerals (“chlorite + kaolinite,” which cannot be analyzed separately) in the aeolian suspension over the Arctic seas averaged 50.9% (see Table 2). Among these two minerals, the chlorite is the second in importance (after illite) component of the Arctic aerosols and bottom sediments. In a series of generalizing papers,^{40,101,103} it was shown that illite and chlorite are most typical for the clay minerals of land soils and bottom sediments of cold and temperate zones. Their distribution corresponds to the climatic zoning, which represents the aeoliation core zoning on land.

In the aeolian material collected during the SPASIBA-91 mission, only trace amounts of minerals of montmorillonite group were found. In the aerosols sampled during the 49th trip of the RV *Dmitrii Mendeleev* in the Kara Sea, the mean content of montmorillonite was only 0.7% of the sum of clay minerals (see Table 2). However, it was much higher (7.4%) in the samples collected in August–October 1994 in the 9th trip of the RV *Professor Logachev* near the Franz Josef Land (see Table 2, Fig. 2). The distribution of montmorillonite (smektite) in the Arctic aerosols is determined by the abundance of basalt (Franz Josef Land, Patomsk Highland, Alaska, basins of the rivers Lena, Khatanga, and Yenisei).

In the soil of the Lena banks, the content of montmorillonite is less than 10% of the sum of all minerals.²⁴ The qualitative estimates of the mineral composition of soils in the Arctic tundra of Eurasia also show that the content of montmorillonite in the surface soil layer is usually low.¹⁰⁴ The high content of smektite was noticed in the surface layer of bottom sediments near the Franz Josef Land.^{105,106} The most part of these islands is typical plateaus from 50 to 600 m high armored by dolerite intrusions and basalt, forming smektite in the process of aeoliation.¹⁰⁷

2.5. Chemical composition

Chemical composition of aerosols is determined by their sources and the types of chemical transformation the aerosol matter is subjected to.⁶¹ Based on the current methods of analysis, we succeeded in studying the chemical composition of both individual aerosol particles and the samples as a whole collected by netting (water-insoluble particles) and air filtering (all aerosol particles larger than 0.5 μm), including sea salts. The chemical composition, like the mineral one, is one of the most important markers of aerosols.

a. Chemical composition of individual aerosol particles

The element composition of individual aerosol particles in the samples collected by netting during the SPASIBA-91 mission was studied with an X-ray microanalyzer at the Chemical Department of the Antwerp University.^{22,27,58}

Table 3. Results of X-ray analysis of individual aerosol particles composition. The aerosol was sampled during SPASIBA-91 mission in August–September of 1991 (Ref. 58)

Sample	Group	Abundance, in %	Mean diameter, in μm	Main elements	Identification
1	1	41.0	1.8	Si	quartz
	2	32.0	1.7	Si, Al, Fe, K	aluminosilicates
	3	11.0	1.9	Fe, Si	Fe–Si-rich
	4	4.0	1.0	Si, Fe	Si–Fe-rich
2	1	43.5	1.7	Si, Al, Fe, K	aluminosilicates
	2	18.3	2.2	Si	quartz
	3	16.5	0.6	Cl, K, Si	fly ash
	4	10.3	1.2	Fe, Si, Cl, K	fly ash
3	1	32.0	1.7	Si	quartz
	2	28.8	1.8	Si, Al, Fe, K	aluminosilicates
	3	10.0	1.3	Fe, Si	Fe–Si-rich
	4	4.3	2.7	Si, Fe, Ca	Si–Fe–Ca-rich
4	1	34.0	1.5	Si	quartz
	2	21.8	1.7	Si, Al, Fe	aluminosilicates
	3	17.5	2.2	Si, Fe	Si–Fe-rich
	4	4.0	1.1	Si, Ti, Al, Fe	fly ash
6	1	35.0	1.5	Si, Al, Fe, K	aluminosilicates
	2	12.8	1.3	Si	quartz
	3	12.5	1.3	Si, Fe	Si–Fe-rich
	4	8.0	1.2	Fe, Si	Fe–Si-rich
9	1	22.7	1.8	Si, Al, Fe, K	aluminosilicates
	2	15.8	2.0	Si, Fe, Al, K	Si–Fe-rich
	3	9.7	2.4	Si, Fe, Mg	Si–Fe-rich
	4	7.9	1.5	Fe, Si, Al, K	Fe–Si-rich

Biogenic particles and black carbon do not give main induced signals in the X-ray region, therefore, only the particles emitting X-ray radiation in response to e-beam bombardment were analyzed in the automatic mode (Table 3).

After the data were processed statistically, four main types of particles were recognized in the Arctic aerosol: (1) aluminosilicate, (2) quartz, (3) Si–Fe- or Fe–Si-enriched particles, (4) combustion spheres (carbon-less ash) enriched in Fe, P, Ni, and some microelements.

Grains of aluminosilicate from 1 to 5 μm in diameter (on average, 1.7 μm) have irregular shape and different degree of rolling. The X-ray spectrum of one aluminosilicate particle is shown in Fig. 8a. Such the particles can be considered as soil particles transported by wind.

Quartz grains in aerosols usually have sizes from 1 to 3 μm and nearly spherical shape. Si–Fe- and Fe–Si-enriched particles are most probably the quartz grains covered with ferrum hydroxide film (so-called river quartz). The variations of the aluminosilicate content in the samples (see Table 3) are likely connected with variability of soil composition.

The combustion spheres enriched in Fe, P, Cl, and microelements had the sizes from 0.3 to 3 μm . The combustion spheres enriched with such elements as Fe, Ni, Cu, Cr, Zn, Ca, Mn, and Sb were detected at manually controlling the microanalyzer. For example, a 0.5- μm sphere containing 52% Sb, 29% Si, and 19% Fe (taking into account only the elements producing the main X-ray signals) was found in the sample 1. Figures 8b and c show the characteristic X-ray spectra of combustion spheres from the Arctic aerosols. The highest concentration of the combustion spheres enriched with microelements was noticed in the sample 16 taken from the air mass arrived from the Norilsk region.

The qualitative element analysis of individual aerosol particles collected by netting in the Laptev Sea in late July of 1995 (ARK XI/1 trip of the ice-breaker *Polarstern*, samples 1 and 2) and in the Kara and Barents Seas in September–early October of 1993 (the 49-th trip of the RV *Dmitrii Mendeleev*, samples 16–18, 20, and 23) was conducted with the SEM-515 scanning electron microscope supplemented with the EDAX PV9900 X-ray microanalyzer (Philips, USA), at the Alfred Wegener Institute of Polar and Marine Research (Bremerhafen, Germany).²⁹ It was shown that the composition of 110 combustion spheres with diameters from 1 to 10 μm varies from region to region (Table 4). They mainly consist of Si, Al, K, Fe, and Mg in the Laptev Sea; Si, Al, Fe, and K in the central part of the Kara Sea; Fe, Ni, Si, and Cu in the southern Kara Sea (income of aerosol from Norilsk); Si, Al, Fe, and Ni in the southern Barents Sea (influence of industrial enterprises of the Kola Peninsula).

Thus, the study of the element composition of individual insoluble aerosol particles in the surface

atmospheric layer over the Arctic seas with the electron microanalyzer has shown that the inorganic portion of the aerosol consists mostly of mineral particles (predominantly, quartz and aluminosilicate) of land soils. Almost all the samples contain small amounts of anthropogenic ash particles enriched in Fe, Ni, Cu, Cr, Zn, Ca, Mn, and Sb. The content of ash particles is a marker of aerosols from industrial regions: Norilsk and Kola Peninsula.

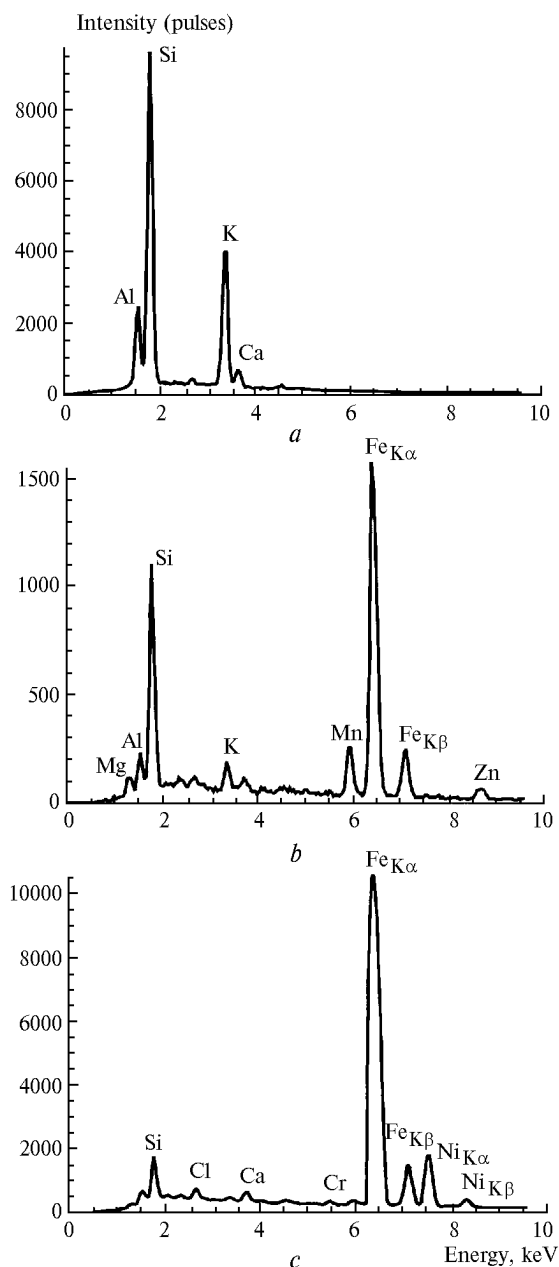


Fig. 8. X-ray spectra of aerosol particles collected by netting during the SPASIBA-91 mission (microprobe analysis was performed at the Antwerp University, Belgium): angular aluminosilicate particle (sample 6, particle size of 5 μm) (a), combustion spheres [sample 1, diameter of 0.7 μm (b), sample 6, diameter of 0.5 μm (c)].

Table 4. Qualitative element composition of combustion spheres of 1 – 10 µm size in aerosol samples, collected by netting in the Laptev, Kara, and Barents Seas in September 1993 (49-th trip of the RV *Dmitrii Mendeleev*) and in July 1995 (ARK-XI/1 trip of the ice-breaker *Polarstern*), and analyzed with the electron microanalyzer EDAX PV9900 at the Alfred Wegener Institute of Polar and Marine Research, Bremerhafen, Germany²⁹

Region, samples	Type of particles by dominant chemical elements
Laptev Sea, samples ARK-XI/1-1* and ARK-XI/1-2	1) Si, Al, K, Fe (Mg, K) 2) Si, Al, Fe (K, Mg) 3) P, Si, Al, S (Mg, Fe, Ca) 4) Fe, Al, P (Cu)
Southern part of Kara Sea, samples DM-49-16, ** DM-49-18, and DM-49-20	1) Fe, Ni, Si (Cu, Al) 2) Fe, Si, Ni (Cu, Al, Mg, Ca) 3) Si, Fe, Cu, Al 4) Si, Al, K (Fe) 5) Fe, Cu, Ni, Si
Central part of Kara Sea, sample DM-49-17	1) Si, Al (K, Fe) 2) Fe, Si, Al 3) Fe, Ni, Si (Cu)
Southern part of Barents Sea, sample DM-49-23	1) Si, Al (Ca, Fe, K, Mg) 2) Fe, Si, Ni (Al, Cu, Mg, Ca) 3) Fe, Ni, Si (Cu, Ca, Al)

* Sample 1 collected in ARK XI/1 trip of the *Polarstern*.

** Sample 16 collected in the 49th trip of the RV *Dmitrii Mendeleev*.

b. Mean element composition of aerosols (insoluble part)

Organic carbon (measured in a sample after its processing by a solution of hydrochloric acid (in proportion 1:1)) is of primary importance in the arctic aerosol sampled by netting.¹⁰⁸ Because of very small amount, the content of C_{org} was successfully determined only in 18 samples collected in the SPASIBA-91 mission, the 49th trip of the RV *Dmitrii Mendeleev*, and in the 15th trip of the RV *Professor Logachev*. It varied from 7.54 to 48.9%. Being recalculated to the organic matter (OM), it makes about 14–97%, i.e., the arctic aerosol is an important source of organic matter. The mean content of C_{org} was 30.2% at standard deviation of 12.5%; this amounted about 60% of the OM. The numerous recent studies show that the organic matter is one of the main components of aerosol above both land and ocean. The content of C_{org} can sometimes achieve 60% of the total mass of a particle.^{98,109–111}

In four samples collected during the SPASIBA-91 mission, E. Cachier (Center of Radioactivity Research, France) determined the concentration of ash carbon. The ratio of ash carbon concentration to that of total carbon in our samples varied from 0.22 to 0.27, what is

typical of industrial aerosols. Earlier, the high concentration of ash carbon was detected at the Alaska coast (Point Barrow) with the ash-to-total carbon ratio achieving 0.40 (Ref. 112). The probable sources of ash carbon are the anthropogenic activity and forest fires. Recently, it was shown that the ash aerosol consisting of submicron particles can be transported far from its source.^{70,113,114} It heavily affects the Arctic ecosystem due to its influence on atmospheric optics at the sacrifice of strong absorption of solar radiation. On the whole, the distribution of ash carbon in the Arctic atmosphere significantly varies in time and is highly inhomogeneous (spotty)^{115–117} due to spotty arrangement of its sources.

The mean content of Si in the netted samples is 13.4% at standard deviation of 7.7%, and the mean content of Al is 2.66% at standard deviation of 1.76% (as analyzed by A.B. Isaeva). Using Al as an indicator of the terrigenous matter and assuming that its mean content in the Earth's crust is 8.23% (Ref. 118), we have determined the content of the terrigenous component in the Arctic aerosols. It averages 32.3%. Thus, the biogenic part makes 67.7%, that is, twice as large. This corresponds to the results of microscopic studies and determination of C_{org} and OM. In the humid equatorial zone of the Northern Atlantic, the terrigenous matter makes about 100% of water-insoluble aerosols at the C_{org} less than 3% (Refs. 57 and 119). The mean value of the Si/Al ratio for the Arctic aerosols, according to our data, is 4.01, what is somewhat higher than that for the Earth's crust (3.42) (Ref. 118) and continental clay (3.06) (Ref. 120), but far higher than the values characteristic of the humid equatorial zone of the Northern Atlantic [2.47 (Ref. 57) and 2.03 (Ref. 119)]. Enrichment of tropical aerosols with Al is likely connected with the fact that the terrigenous sources in this case are the aeolian cores and soils of the tropical zone more rich in Al relative to Si (Ref. 121).

The content of chemical elements in the netted samples varies very widely. To determine the origination of aerosols, the enrichment coefficients were calculated relative to the mean composition of the Earth's crust. For each element, the enrichment coefficient (EC) was calculated as

$$EC = (El./Al) \text{ sample} / (El./Al) \text{ Earth's crust},$$

where El. and Al are the concentrations of the element of interest and aluminum in the sample and in the Earth's crust,¹¹⁸ respectively. For example, in the netted samples of the SPASIBA-91 mission, the considered elements were divided into two groups in accordance with the EC value:

(1) the elements typical of the Earth's crust ($EC < 10$): Na, Mg, Al, K, Ca, Sc, Mn, Fe, Co, Ni, As, Rb, Cs, Ba, rare-earth elements, Hf, Ta, Th, U;

(2) the anthropogenic elements ($EC > 10$): Cr, Cu, Zn, Se, Br, Ag, Cd, Sb, W, Au, Pb.

When nets were washed by bi-distilled water, sea salt was removed, therefore, the group of elements connected with sea salt had no effect. The highest ECs of aerosols for Cr, Cu, Sb, and Pb were observed in the sample 6 taken above the Laptev Sea (at the southerly wind) (see Fig. 2). Even for nickel, which is usually classified as a crust element, the EC in the samples 5 and 6 was more than 10 (12 and 13.5, respectively). The samples 5 and 6 were also characterized by the largest amount of the anthropogenic combustion spheres found by the electron probe analysis. Obviously, the main source of pollution of these samples was in the region of Norilsk according to the data of electron probe analysis and EC analysis.

The Norilsk Mining-Metallurgical Plant is one of the world's largest plants on mining and processing of copper-nickel ore. This plant is one of the main sources of pollution in the north-eastern part of Russia emitting the compounds of S, Se, Cu, Ni, Co, Sb, As, Pb, Zn, and Hg into the atmosphere.^{122–124}

The sample 16 taken on the September 18, 1993, in the 49th trip of the RV *Dmitrii Mendeleev* in the Yenisei gulf (see Fig. 1) at strong (10.4 m/s) southerly wind had the highest, as compared to other samples, coefficients of enrichment for such elements as Se (EC = 22 000), Ni (418), and As (43). The highest content of combustion spheres was also detected in the same sample taken along the path of air masses from Norilsk (see Fig. 7). This confirms the earlier idea that the Norilsk region is the significant source of pollution of the Arctic atmosphere,^{10,125} especially, above the Laptev and Kara Seas.

Earlier it was shown that the natural and anthropogenic sources of selenium in aerosols were almost equal.^{126,127} The most important natural process delivering selenium to the atmosphere is generation of its organic compounds by phytoplankton, because the biochemical behavior of Se is close to that of S (Refs. 128 and 129). The main anthropogenic source of Se is the combustion of coal, oil products, and wood, as well as mining and metallurgic plants (especially, copper production).^{126,127} Enrichment of aerosols with selenium is one of the markers of functioning of copper-nickel plants.

In August–October of 1994, aerosols were studied over the Barents Sea and the St. Anna Channel in the 9th trip of the RV *Professor Logachev*.^{25,26} Twenty samples have been collected by netting (see Fig. 2). The highest coefficients of enrichment for Co, Ni, Cu, and Zn were observed in the sample 17 taken near Murmansk. That sample contained the anthropogenic combustion spheres in a large amount. The content of most chemical elements in the samples of aerosols netted in this trip was lower than their mean content in the Earth's crust, what can be explained by "dilution" of insoluble aerosols with the organic matter. The content higher than in the crust was noticed only for Cr, Cu, Zn, As, Se, Br, Ag, Sb, and Au; the main source of these elements is anthropogenic. The

comparison of mean composition of the samples near the Franz Josef Land (samples 3–10) and near the Kola Peninsula (samples 15–19) (see Fig. 2) has shown that the content of chromium was roughly 1.5 times higher than that near the Franz Josef Land, whereas the content of nickel was roughly five times higher than that near the Kola Peninsula. The elevated content of Cr was observed in aerosols near the Franz Josef Land because the main magmatic rocks – dolerite and basalt¹⁰⁷ enriched with chromium^{118,130} were abundant there. The increase of the nickel content near the Kola Peninsula is explained by the income of aerosols polluted by emissions of the metallurgic plants of the Kola Peninsula. The spectra of aerosol enrichment with metals are very close for the Norilsk region and the Kola Peninsula, because the plants situated on the Kola Peninsula use mostly the ore from Norilsk.

To reveal the geochemical relations between the elements, the correlation and factor analyses were performed. The correlation matrix of the element composition for the netted samples of the aerosols over the Kara Sea is given in Table 5. For 18 samples the critical level of significance of the correlation coefficient at the 5-% level is 0.468 (Ref. 131). Very high positive (> 0.87) correlation coefficients are between the terrigenous origin of a sediment) and Na, Ca, Fe, Cs, Ag, Hf, Ba, Th. The main sources of these elements in the water-insoluble fraction of aerosols of the Kara Sea are the soils of tundra and forest-tundra, as well as the terrigenous aeolian material from more remote regions, which is transported to the Arctic by air masses (the influence of sea salt is excluded at processing the samples). The lower, but still significant, correlation coefficients are between Sc and Cr (0.65) and Sc and Mo (0.57), whereas the correlation coefficient between Cr and Mo is 0.99; chromium also has high correlation coefficients with Br (0.89), Ag (0.88), Ca (0.85), Au (0.75), Fe (0.74), Zn (0.74), Ba (0.73), and Th (0.70). We can assume that the main sources of Cr and Mo also are soil and surface rocks. Very high positive correlation coefficients (from 0.94 to 0.99) are noticed between Ni, Co, and Se, the main source of which is the smoke of the Norilsk Metallurgical Works.

To reveal the groups of genetically related elements, we used the factor analysis.¹³² In this analysis, the content of any element in each sample is considered as a result of the sum action of hypothetical sources (factors). The results of the factor analysis of the samples netted in September 1993 in the Kara Sea are given in Table 6.

The first factor makes the main contribution to the total dispersion; it is equal to 77.2%. The load of Ni on this factor is abnormally high (8.63), whereas the loads of other elements are close to zero. The first factor represents the contrast role of the source of nickel as compared to the sources of other elements. This dominating source of nickel determining its biochemical behavior in the Arctic aerosols is, as was noted, the anthropogenic delivery from the Norilsk region.

Table 5. Correlation matrix of chemical composition of netted samples of aerosol over the Kara and Barents Seas (September 1993, the 49th trip of the RV *Dmitrii Mendeleev*); $n = 18$ samples, the critical level of the correlation coefficient at the 5-% significance level is 0.468

	Na	Ca	Sc	Cr	Fe	Co	Ni	Zn	Se	Br	Mo	Ag	Sb	Cs	Ba	Hf	Ir	Au	Th
Na	1.00	0.81	0.99	0.59	0.96	0.22	0.41	0.31	-0.05	0.22	0.48	0.89	-0.11	0.99	0.88	0.98	-0.01	0.36	0.98
Ca		1.00	0.88	0.85	0.90	0.02	-0.52	0.68	-0.20	0.64	0.93	0.80	0.12	0.89	0.96	0.89	0.00	0.77	0.90
Sc			1.00	0.65	0.98	0.23	0.45	0.35	-0.05	0.30	0.57	0.91	-0.10	0.99	0.93	0.99	0.01	0.48	0.99
Cr				1.00	0.74	0.05	-0.41	0.74	-0.10	0.89	0.99	0.88	0.12	0.65	0.73	0.67	0.13	0.75	0.70
Fe					1.00	0.31	0.73	0.42	0.04	0.40	0.70	0.89	-0.06	0.99	0.94	0.98	0.01	0.52	0.98
Co						1.00	0.99	-0.16	0.94	-0.10	0.46	0.06	-0.28	0.42	0.42	0.21	-0.06	-0.01	0.20
Ni							1.00	-0.53	0.97	-0.51	-0.36	0.23	-0.56	-0.35	-0.04	-0.09	-0.12	-0.33	-0.12
Zn								1.00	-0.26	0.68	0.89	0.81	0.15	0.34	0.46	0.37	0.06	0.46	0.38
Se									1.00	-0.11	0.14	0.06	-0.23	-0.01	-0.01	-0.07	-0.18	-0.07	-0.06
Br										1.00	0.87	0.85	0.22	0.28	0.43	0.34	0.14	0.82	0.36
Mo											1.00	0.92	0.06	0.57	0.84	0.62	0.98	0.69	0.63
Ag												1.00	0.20	0.93	0.81	0.85	0.80	0.75	0.91
Sb													1.00	-0.01	-0.16	-0.10	-0.10	-0.15	0.16
Cs														1.00	0.93	0.99	-0.02	0.43	0.99
Ba															1.00	0.95	-0.06	0.64	0.95
Hf																1.00	0.03	0.50	0.99
Ir																	1.00	0.06	0.01
Au																		1.00	0.51
Th																			1.00

Table 6. Matrix of factor representation of element composition of samples of aerosol ($> 1 \mu\text{m}$) netted in the Kara Sea in September 1993 in the 49th trip of the RV *Dmitrii Mendeleev* ($n = 18$ samples)

Factor	I	II	III	IV	V
Part of total dispersion	77.2%	11.7%	3.2%	2.3%	1.9%
Na	0.05	0.86	-0.45	0.04	-0.16
Ca	-0.06	0.95	-0.03	0.25	-0.19
Sc	0.05	0.90	-0.40	-0.01	-0.14
Cr	-0.05	0.86	0.25	-0.20	-0.01
Fe	0.08	0.93	-0.33	-0.07	-0.06
Co	0.12	0.19	-0.28	-0.05	-0.42
Ni	8.63	0.01	0.03	-0.01	-0.02
Zn	-0.07	0.70	0.90	-0.47	-0.25
Se	0.12	-0.04	-0.20	0.08	0.59
Br	-0.06	0.63	0.55	-0.10	0.04
Mo	-0.04	0.96	0.51	-0.41	0.76
Ag	0.03	1.23	0.46	1.25	0.16
Sb	-0.07	0.02	0.48	0.14	-0.68
Cs	-0.04	0.90	-0.43	0.01	-0.12
Ba	-0.01	0.93	-0.30	-0.18	0.04
Hf	-0.01	0.91	-0.38	-0.06	-0.13
Ir	-0.01	0.15	0.16	0.09	0.16
Au	-0.04	0.67	0.25	-0.11	-0.05
Th	-0.01	0.92	-0.36	-0.03	-0.13

The contribution of the second factor is equal to 11.7%. The high positive load (> 0.62) on this factor is given by Na, Ca, Sc, Cr, Fe, Zn, Br, Mo, Ag, Cs, Au, Th. The main source of these elements in the water-insoluble fraction of aerosols is the terrigenous material from the land soil and rocks. Such elements as Se, Ni, Co, and Ir have the lowest effect on the second factor. The main source of Se, Ni, and Co is the material transported by air masses from the Norilsk region. In the geochemical behavior, cobalt is close to nickel,¹³⁰ and the content of cobalt in copper-nickel ore of the Norilsk region is relatively high.¹³³ The content of

selenium in the copper-nickel ore and emissions of copper-nickel plants is very high, as was noted above.

The contribution of the third factor is equal to 3.2%. The positive load (> 0.47) on this factor is given by Zn, Br, Mo, Ag, and Sb, that is, the elements which can be accumulated in vegetable fibers,¹³⁰ and the high negative load is given by Na, Sc, Fe, Cs, Ba, and Th, that is, the elements typical of the mineral phase.

The contributions of the fourth and fifth factors are insignificant (2.3 and 1.9%). The difference in loads of the elements on these factors may be connected with the variety of regions, from which the crust and anthropogenic aerosols come.

On the whole, the content of most chemical elements (Na, Al, K, Ca, Sc, Fe, Co, Rb, Zr, Cs, Ba, rare-earth elements, Hf, Ta, Th, U) in the insoluble fraction of the Arctic aerosols is lower than the mean value for the Earth's crust. The content of Cr, Cu, Zn, As, Se, Br, Ag, Sb, and Au is far higher than the mean value for the Earth's crust; their main source is anthropogenic. The most significant sources of aerosol pollution in the Russian Arctic are mining and metallurgical works of Norilsk and the Kola Peninsula, as well as forest fires; episodically (especially, in winter) the influence of the far transport of pollutants from Europe, Asia, and North America is tangible.

c. Element composition of aerosols (taking into account sea salt; filtering method)

The element composition of aerosols as a whole was studied simultaneously in the samples obtained by filtering and netting. In the case of filtering, the composition of particles larger than $0.5 \mu\text{m}$, including (in contrast to the netted samples) particles of sea salt, was examined.

In the Laptev Sea in July–September of 1995 (Refs. 63 and 134), the highest content of Na

(1230 ng/m³), the marker of sea salt, was observed, when the vessel moved in the open water at rather high speed of wind (on average, 11.8 m/s, sometimes up to 16 m/s). The content of Sc (the marker of continental matter) in this sample was also relatively high (0.009 ng/m³), but the sea component prevailed. The coefficient of enrichment for Na relative to the mean content in the Earth's crust is very high (135). The contribution of sea salt particles to formation of aerosol composition in this region is very significant, as was shown earlier for other ocean regions.⁷⁰

The highest content of Sc (0.023 ng/m³) at the relatively low content of Na (410 ng/m³) was observed in August, 5, in the sample 6; EC(Fe) and EC(Zn) for that sample were relatively low, and the content and EC for Cr and As were high. This indicates that aerosols here are terrigenous and polluted with Cr and As. Measurements of the aerosol particle size distribution have shown that the content of particles larger than 0.5 μm was relatively high (11.3 cm⁻³). The synoptic situation a day before was characterized by income of polar air masses from the north to the region of Khatanga through the Kara Sea and Taimyr Peninsula; on 06:00 GMT of August, 5, the surface center of low pressure was located under the Northern Land. During sampling, the strong (from 13.5 to 15 m/s) south-easterly wind was observed. Apart from the land source, the ice located near the sampling site and rich in sedimentary material was likely one more source of mineral aerosol particles, as was demonstrated earlier for other regions.¹³⁵ The ice rich in sedimentary material is abundant in the Arctic.^{136–138} In the ARK XI/1 trip, the *Polarstern* crossed a large field of "dirty" ice. According to the NOAA AVHRR data, late in July of 1995 the area covered by dirty ice only in the Laptev Sea was 24 000 km² (Ref. 139).

The maximum content and EC of anthropogenic elements in aerosols of the Laptev Sea were observed in the sample 4. The content of both terrigenous (Sc, 0.0016 ng/m³) and marine (Na, 240 ng/m³) matter in that sample was low as compared to other samples, whereas the ECs for Fe, Co, Cr, and Zn were the highest in this mission. During the sampling the weak southerly–south-easterly wind (5.5 m/s) was observed. The content of aerosol particles larger than 0.5 μm was low (1.26 cm⁻³). The low contents of Sc, Na, and aerosol particles larger than 0.5 μm indicate that the anthropogenic elements are contained mostly in the fine aerosol fraction. There exist two most probable sources of fine particles: (1) generation of salt aerosol particles at destruction of bubbles in the surface microlayer of the sea water; (2) far transport of fine particles from regions with the open surface of the sea water. The coefficient of enrichment for Na in the sample 4 was high and indicative of predominantly marine origination of these aerosols. As has been noted earlier, sea salt particles are enriched with many microelements because of fractionation in the surface microlayer of the sea water.^{62,70}

The mean contents of chemical elements over the Laptev Sea are compared with the literature data in Table 7. The mean values for Na (494 ng/m³) are close to the content of Na in aerosols of the Fram Strait near the edge of an ice field¹³⁵ and much higher than that in the Central Arctic covered by many-year ice,¹³⁵ in Arctic islands,^{11,78} and in the Antarctic.¹⁴⁰ The main cause of the high content of Na and other elements of marine aerosols is that the sampling was performed in the Laptev Sea in summer, when a significant part of water surface was open.¹⁴⁶

According to our data, the mean content of Sc in aerosols of the Laptev Sea is 0.0081 ng/m³, what is higher than in some other regions of the Arctic and Antarctic (see Table 7). The increased content of scandium, which is typical terrigenous element, is connected with the closeness of the land free of snow at the time of measurement and rarely with re-suspension of the mineral material from dirty ice, as well as with income of products of oil and coal burning.

The content of chemical elements in aerosols of the Kara Sea in September, 1993, was close to their content in other Arctic regions or lower (see Table 7). It is far (by one to two orders of magnitude) lower than usually in the Arctic in the late winter – early spring.^{147–148} And it is one to two orders of magnitude lower than that over the Black and Mediterranean Seas (influence of the arid zone and industrial impact).^{145,150,151} Thus, the air in the Arctic region under study can be considered relatively clean. The content of microelements in the Antarctic air is even lower,^{140,152} what is explained by remoteness of the Antarctica from most sources of natural and anthropogenic aerosol.

As for the netted samples, the high content of Se in the samples on Whatman-41 filters and high EC were measured in the samples 7–9 (Yenisei Gulf at air masses coming from Norilsk).²² In those samples the high EC was also measured for Co. These results, along with the data of scanning electron microscope and electron probe analysis, as well as the element composition of insoluble aerosols are indicative of significant pollution of sea aerosols even at a far (> 300 km) distance from Norilsk.

As an additional indicator of anthropogenic aerosols, we have determined the ratios of tracer elements (Zn/Sb, As/Sb, Se/Sb) characteristic of different sources. The use of these ratios was proposed by Rahn and Lowenthal.^{153,154} When using the method of back trajectories of the air mass transport,^{11,78,155} the sources can be separated, as well as their specific contribution to the aerosol composition. The values of Zn/Sb and Se/Sb, obtained by us for aerosols of the Kara Sea, are by an order of magnitude higher than those available in the literature for aerosols of the Norilsk region. One of the main causes of this difference is likely insufficiently reliable data on the element composition of aerosols of the Norilsk region, especially for Sb, Se, and As.

Table 7. Content of chemical elements in aerosol of Arctic and other regions, ng/m³ (mean values)

Region, time	Number of samples, <i>n</i>	Na	Ca	Sc	Cr	Fe	Co	Ni	Zn	As	Se	Br	Rb	Sb	Source
Laptev Sea, July–September 1995	12	494		0.0081	3.97	142	0.102		15.4	0.38		1.74			This work
Kara Sea, September 1993	14	24.9		0.00022	0.044	2.81	0.0041		1.58		0.049			0.0032	Ref. 22
St. Anna Channel and Barents Sea, August–October 1994	10	1760	283	0.0033	5.66	135	0.30	8.2	43	0.15		0.1		0.064	Ref. 26
Norwegian and Greenland Seas, July–August 1996	5	1517	155	0.026	0.665	800		19.1	34.2	0.64	0.28	0.5	0.87	0.098	This work
Fram Strait, June–July 1997	8	415	9.2		0.56	57		11.1	18.8	0.08	0.3	0.044	1.11	0.16	This work
Central Arctic, August–October 1991	7	20	3	<0.003	<1.1	5.9	0.019	<0.18	0.18	0.022	<0.2	0.25	0.042	0.019	Ref. 135
Fram Strait, August–October 1991	3	490	23	<0.004	<1	10.6	0.014	<0.2	0.19	<0.06	0.088	1.26	0.052	<0.005	Ref. 135
Northern Land, April–May 1985, 1986, and 1988	22	90	180	0.014	1.6	30	0.075	6.2	5.6	1.8	0.3	4.1		0.19	Ref. 11
Spitsbergen, Ni-Alesund, summer 1984	13	66	7.3	0.0012	0.56	5.6	<0.004	<0.2	<0.15	0.01	0.035	0.73	<0.08	0.0024	Ref. 78
Spitsbergen, Ni-Alesund, winter 1983, 1984, and 1986	46	230	34	0.0043	<0.4	17.8	0.0096	0.29	3.9	0.52	0.156	8.8	0.083	0.092	Ref. 78
Canadian Arctic, st. Alert, March 1985	12	234.5	62.6	0.01		50	0.032		8.5	0.349	0.081	18.5		0.06	Ref. 141
Alaska, Point Barrow, March–April 1986	280	240	53			38		4	11			25			Ref. 142
Greenland, st. Summit (3170 m), June–August 1990	29	4.3	16	0.003		9.8	0.011		1.5	0.029	0.02	0.95			Ref. 143
North Sea, Helgoland Island, 1986–1990	85								99	8	2.4			1.8	Ref. 144
Black Sea, western part, June–September 1988	14	1400		0.08	9	420	0.25	4.9	46	1.1	0.73	0.02	1.6	0.6	Ref. 145
Antarctic		3.3		0.00016	0.04	0.62	0.00049		0.033						Ref. 140
Region, time	Number of samples, <i>n</i>	Cs	Ba	La	Ce	Nd	Sm	Eu	Yb	Lu	Hf	Au	Th	U	Source
Kara Sea, September 1993	14											0.0029			Ref. 22
St. Anna Channel and Barents Sea, August–October 1994	10	0.016	0.89	0.064	0.11	0.035	0.0091	0.0021	0.0026	0.0004		0.0026		0.041	Ref. 26
Norwegian and Greenland Seas, July–August 1996	5	0.032	3.39	1.94	2.49	0.46	0.07	0.017	0.026	0.004	0.088		0.046	0.53	This work
Fram Strait, June–July 1997	8	0.023	2.03	0.21			0.011	0.0018	0.009	0.0012	0.01		0.0083	0.21	This work
Central Arctic, August–October 1991	7	<0.04	<4	<0.013	<0.3		0.0005	<0.01		<0.006			<0.03		Ref. 135
Fram Strait, August–October 1991	3	<0.04	<5	<0.03	<0.3		<0.003	<0.016		<0.007			<0.035		Ref. 135
Northern Land, April–May 1985, 1986, and 1988	22						0.01				0.064	0.066	0.018		Ref. 11
Spitsbergen, Ni-Alesund, summer 1984	13	0.003	<0.7	0.0055	<0.02		0.00072	<0.001		<0.0004			0.0017		Ref. 78
Spitsbergen, Ni-Alesund, winter 1983, 1984, and 1986	46	0.0089	<1.5	0.0137	<0.05		0.003	0.00116		<0.0028			0.0037		Ref. 78
Canadian Arctic, st. Alert, March 1985	12			0.029	0.047		0.004						0.0042		Ref. 141
Black Sea, western part, June–September 1988	14	0.17	4.5	0.37	0.74		0.04	0.01	0.02				0.11		Ref. 145

It is very interesting to compare the mean values of EC for the aerosols sampled by netting and filtering through the Whatman-41 filters (Table 8). It is seen that for most elements the EC of the filtered samples is one to two orders of magnitude higher than that for the netted samples. This proves that such elements are largely connected with submicron particles, which hardly can be trapped by nets. The only exclusion is Co, for which the mean values of the EC are almost the same for the both sampling methods. It is proved above that the combustion spheres from 0.2 to 10 μm in size can be enriched with some elements, what is also supported by the chemical composition of different aerosol fractions. It is shown that the concentrations of many microelements are higher in the finest fractions.^{156–158} Thus, the aerosol particle size distribution becomes one of the most important factors determining the aerosol chemical composition, what corresponds to the data on the water suspension, cryosols, and sediments.

In the region of the St. Anna Channel and in the Barents Sea, in August–October of 1994 during the 9th trip of the RV *Professor Logachev*, ten samples were collected by the air filtering through the AFA-KhA-20 filters.^{25,26} The contents of almost all elements (except for bromine and copper), measured by us, were higher than those given in Ref. 78 for Spitsbergen (at the distance of about 1 km from the shore). Moreover, our data exceed even spring values obtained in the period of maximum pollution of the Arctic atmosphere.¹¹ Only As, Sb, and Au contents, as well as already mentioned Br and Cu in the Northern Land were higher. The variability of the aerosol element composition was very wide from sample to sample: the values of rms deviations from the mean concentrations were about 100% for almost all elements. In this case, this represents not only temporal, but also spatial variability of the fields of element concentrations, because the sites of sampling changed in accordance with the vessel route.

The enrichment coefficients for most elements in the aerosol over the St. Anna Channel and the Barents Sea (Fig. 9) are close in the order of magnitude to the mean values for the Arctic islands.²⁶

Data on EC (Fig. 9) and results of the two-element correlation analysis allow the chemical

elements to be divided into three groups: (1) the elements characteristic of the Earth's crust (Sc, rare-earth elements, Th); (2) the marine elements (Na and Br); (3) the elements of mixed marine, terrigenous, and anthropogenic origin (As, Ni, Cu, Se, Fe, Co, and Zn). The relatively high ECs of the rare-earth elements in the filtered samples (10–15) are probably connected both with the fact that the aerosols sampled by filtering consist largely of sea salt, which is characterized by high concentrations of these elements,^{62,156} and with the peculiarities of the composition of soils on the land adjacent to the region of observations, in particular, Kola Peninsula.

The high value of the coefficient of correlation between the contents of Br and Na ($n = 9$ samples; $r = 0.978$; the critical value of the correlation coefficient for nine samples is equal to 0.798 at 1-% significance level) indicates that the main source of Br in the Arctic aerosols in the periods of our investigation was sea salt.

The significant enrichment of the aerosol with Ni (see Fig. 9), in our opinion, is connected not only with its atmospheric transport from the regions of Norilsk and Kola Peninsula, but also with its fractionation at carry-over from sea water to the atmosphere.^{62,156} This is indicated by high coefficients of correlation between Ni and Na.

As for aerosols of the Kara Sea (September, 1993), the ECs of the filtered samples from St. Anna Channel and the Barents Sea (August–October, 1994) proved to be higher than those for the netted samples (see Table 8). This is explained by the fact that the filtering holds submicron aerosol particles enriched with a lot of chemical elements, including sea salt.

In the Norwegian and Greenland Seas, as well as in the western Barents Sea, the composition of aerosols was studied in July–August, 1996, during the 15th trip of the RV *Professor Logachev* (see Fig. 1). The mean contents of the typical marine element Na and the typical terrigenous element Sc were high (1517 and 0.0026 ng/m^3 , respectively) (see Table 7). The EC indicates that the main source of most elements is sea salt. However, for Ni, Zn, As, Se, and Sb the main source is anthropogenic. The indicator ratios Zn/Sb and As/Sb show that this source is in Scandinavia.

Table 8. Comparison of enrichment coefficients (EC) of aerosols sampled by netting (insoluble particles larger than 1 μm) and filtering through Whatman-41 filters in the Kara Sea in the 49th trip of RV *Dmitrii Mendeleev* (September, 1993) and AFA-KhA filters in the 9th trip of RV *Professor Logachev* (August–October, 1994)

Method	Parameter	Elements										
		Na	Cr	Fe	Co	Ni	Zn	Se	Br	Mo	La	Au
49th trip of RV <i>Dmitrii Mendeleev</i>												
netting	mean	0.65	9.5	2.79	12	–	101	3430	34	42	–	14000
	standard deviation	0.29	5.4	1.13	18	–	99	5750	20	24	–	11480
filtering	mean	152	62	7.9	15	–	3550	108800	9560	3050	–	43300
	standard deviation	148	51	6.1	15	–	3290	84200	12210	5070	–	35300
9th trip of RV <i>Professor Logachev</i>												
netting	mean	1.1	40	3.9	5.1	6.1	198	–	13	–	2	5750
	standard deviation	1.8	23	1.4	4.7	10.3	170	–	13	–	1	3140
filtering	mean	785	470	16	90	1320	5060	–	420	–	15	5060
	standard deviation	1200	300	8	116	2630	3700	–	680	–	17	6210

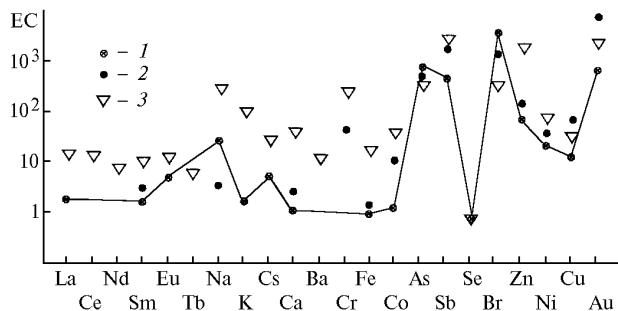


Fig. 9. Enrichment coefficients (ECs) of filtered samples of Arctic aerosol: Barents Sea and St. Anna Channel, August–September, 1994, (the 9th trip of the RV *Professor Logachev*) (1); Northern Land, spring of 1985, 1986, and 1988 (Ref. 147) (2); Spitsbergen, winter, 1983–1984 (Ref. 78) (3).

The mean contribution of air masses of different origin coming to the observation sites located in the Franz Josef Land, the Northern Land, and Wrangel Island is given in Ref. 159 along with the distribution (5 days after) of outgoing air masses. The transport of the matter along the considered trajectories is estimated for six anthropogenic markers with known spatial distribution of emissions.^{160,161} It should be kept in

mind that anthropogenic pollutants can come to the observation sites not only from the continents, but also from the Arctic, where in winter and spring they experience mixing and redistribution due to their long lifetime.⁵⁰

The technique for estimating the contributions of particular industrial regions to content of particular anthropogenic pollutants in the atmosphere above the observation sites was described earlier.⁴⁹ Figures 10 and 11 show the contributions of the source regions to the concentrations of As, Pb, and Cd (Ref. 162). As was shown in Ref. 49, the dependence for Ni and As, V and Pb, Zn and Cd is analogous in pairs, because spatial distributions of the emission are similar. The contribution of each source is proportional to the emission power of a particular pollutant in a particular region and the function, whose values are different for each pair “source – observation site” and determined by synoptic and meteorological parameters of the atmosphere along the air masses path between these sites. Let us underline that the most polluting source may be not the most powerful emitter. This depends on the position of an observation site and on a particular pollutant.

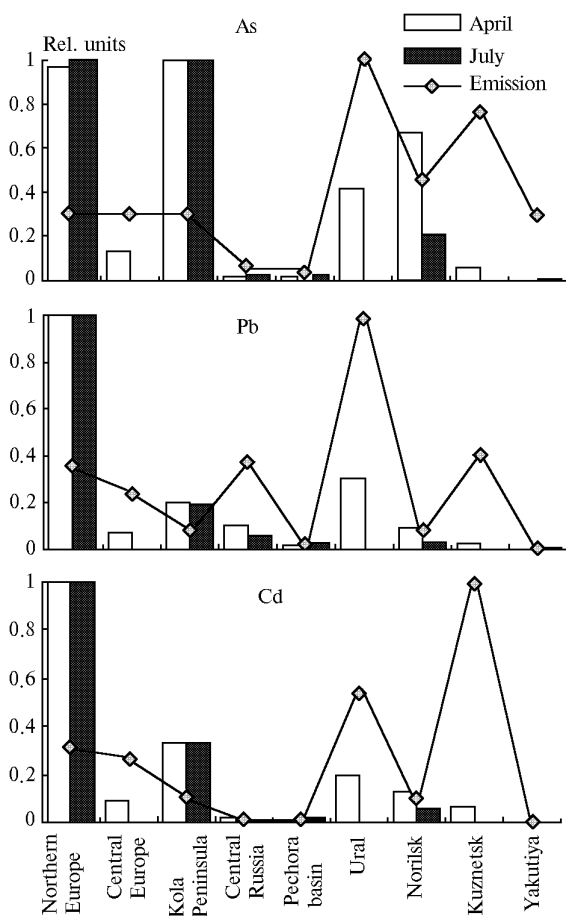


Fig. 10. Contribution of industrial regions to atmospheric pollution with arsenic, lead, and cadmium on the Franz Josef Land, as well as the power of emission of these elements by source regions.¹⁶²

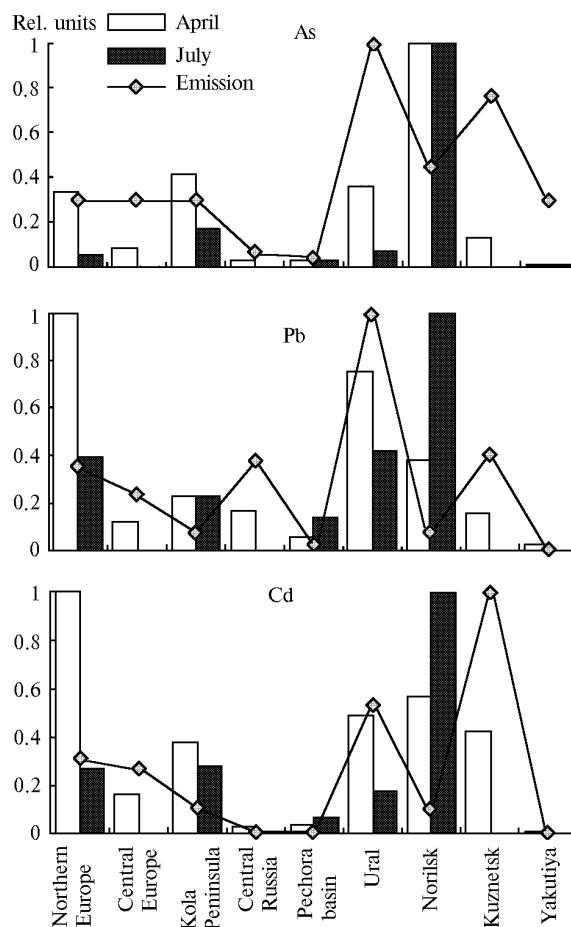


Fig. 11. Contribution of industrial regions to atmospheric pollution with arsenic, lead, and cadmium on the Northern Land, as well as power of emission of these elements by source regions.¹⁶²

Table 9. Mean concentrations of six anthropogenic chemical elements in the atmosphere of the Franz Josef Land and the Northern Land, ng/m³

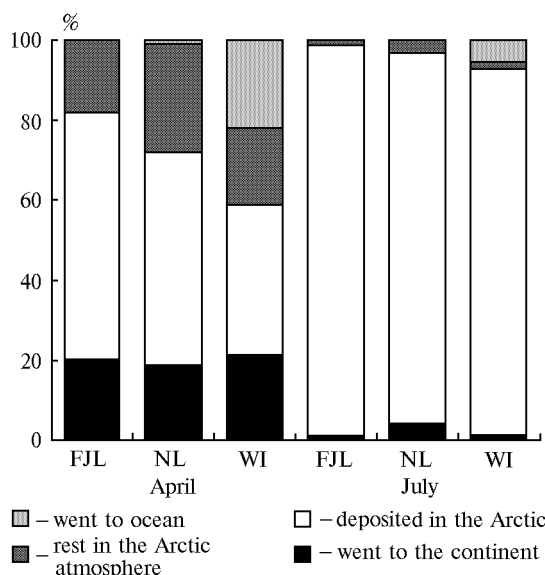
Observation site	Data sources	As	Ni	Pb	V	Zn	Cd
Franz Josef Land	winter–spring, Ref. 163	0.15–0.52	0.1–2	0.5–16	0.09–2.8	2–20	0.12–0.8
	calculated	0.26	0.85	3.3	0.85	1.9	0.08
	data	0.007	0.022	0.09	0.021	0.056	0.002
Northern Land	spring, Ref. 11	0.26–1.8	1.1–11	3.5	0.73–0.8	0.6–13	0.09
	calculated	0.19	0.65	1.9	0.46	1.1	0.048
	data	0.019	0.063	0.1	0.02	0.04	0.0026

As is seen from Table 10, for five days a portion of air (and, correspondingly, pollutants) is transported from the observation sites to the continents or oceans. This can be considered as one of the channels of self-clearing of the arctic atmosphere. The statistical analysis of the spatial distribution of air masses, performed for the first time five days after their passage over an observation site, allowed us to estimate the mean (for each month under consideration) balance of passive pollutants for each site (Fig. 12) (Ref. 159). In spring, only 20% of anthropogenic pollutants, passing over the Russian Arctic, then leaves the arctic regions and deposits on the surface to the south of 70°N. Almost the same amount (18–27%) is left in the arctic atmosphere, redistributing there, what confirms the repeatedly observed phenomena of accumulating and mixing the atmospheric pollutants in the Arctic as in a big reservoir (what is a characteristic of winter and spring seasons). In spring, 22% pollutants move from the region of Wrangel Island to the northern part of the Pacific Ocean, from which they are washed out on the surface in the region of the Arctic front. In summer almost all passive pollutants (more than 90%) passing over the Russian Arctic are then deposited in some or other part of the arctic regions.

Table 10. Distribution of passive pollutants within five day of passage over observation sites, %

Month	Observation site	Went to the continent	Deposited in the Arctic	Rest in the Arctic atmosphere	Went to ocean
April	Franz Josef Land	20	62	18	0
	Northern Land	19	54	27	1
	Wrangel Island	21	38	19	22
July	Franz Josef Land	1	98	1	0
	Northern Land	4	93	3	0
	Wrangel Island	1	92	1	6

The amount of aerosol material in the atmosphere varies widely – for several hours or days it can change by tens and hundreds times. Therefore, such markers as mineral, chemical, and isotopic composition of aerosols, and composition of biogenic residues, rather than the content itself, prove to be more indicative as characteristics of air masses.

**Fig. 12.** Mean balance of passive pollutants after passage over observation sites.

Being recalculated to the air volume, the data on these characteristics also turn to be widely variable. They become more indicative being recalculated to the matter (in percent of the dry aerosol matter). Along with the EC and correlation coefficients, the ratios “element/element-indicator of the terrigenous matter” (most often Al, Sc, Ti) and “element/element-indicator of the sea salt” (often Na, Cl) are recently used for this purpose, as well as a comparison between the groups of elements (minerals) and the correlation analysis. As a result of such processing (for the water suspension and bottom sediments), source provinces, atmospheric provinces, and deponents (main regions of deposition of aerosols of the given composition), etc. can be revealed. The method of element ratios allows revealing the pollution sources. The ratio to Sb proves to be very indicative (Table 11). Using this ratio, we succeeded to find at least 14 regions polluting the atmosphere of the Northern Hemisphere. Thus, the chemical composition, including polluting elements, as well as radioactive isotopes,^{1,164} can serve tracers of sites of origination and transport of air masses (method of elements-markers in addition to the method of minerals-markers), and, conversely, the study of the fields of natural minerals and elements permits prediction of probable distribution of their anthropogenic analogs.

Table 11. Concentration ratios of tracer elements in emissions of some arctic industrial source-regions and other regions (Ref. 11)

Number	Region	Concentration ratios							
		As/Sb	Se/Sb	V/Sb	Zn/Sb	Mn/Sb	Cr/Sb	Ni/Sb	Pb/Sb
I	Kola Peninsula	7.2	1.1	5.3	7.8	4.6	5.3	28	32
II	Pechora basin	0.96	1.8	5.5	4.7	6.2	6.8	6.1	17
III	Norilsk	9.8	1.3	5.2	10.5	1.1	1.2	37	30
IV	Ural	5.9	1.9	32	42	12	15	17	101
V	Yakutiya	1.6	2.2	5.0	6.5	10.5	12	8	32
VI	Kuznetsk	18	2.5	6.6	390	16	19	5.8	160
VII	Donetsk	1.1	2.0	17	44	19	18	8.9	59
VIII	Baikal	5.0	1.5	3.0	8.0	3.5	3.7	4.5	65
IX	Europe without the USSR	12	1.0	77	200	36	39	33	260
X	ETC	35	1.5	140	270	86	89	75	550
XI	Eastern shore of the USA	0.74	1.0	26	28	3.4	–	–	–
XII	Middle West of the USA	2.6	4.4	1.5	46	12	–	–	–
XIII	Great Britain	4.1	0.9	52	87	26	28	22	250
XIV	Scandinavia	24	1.5	220	460	47	49	72	640

Knowing the standard for the Earth's crust ratios $El./Al$ or $El./Sc$, we can determine the content of elements in excess over the standard background value (excess content) in aerosol samples. Such calculations were made for V – the characteristic element of the pollution connected with burning of oil products and coal (Fig. 13).¹⁶⁵ It is seen that the content of V_{exc} in the Arctic is maximum in winter, as well as that the winter fallout has a specific character and clearly differs from the summer one.

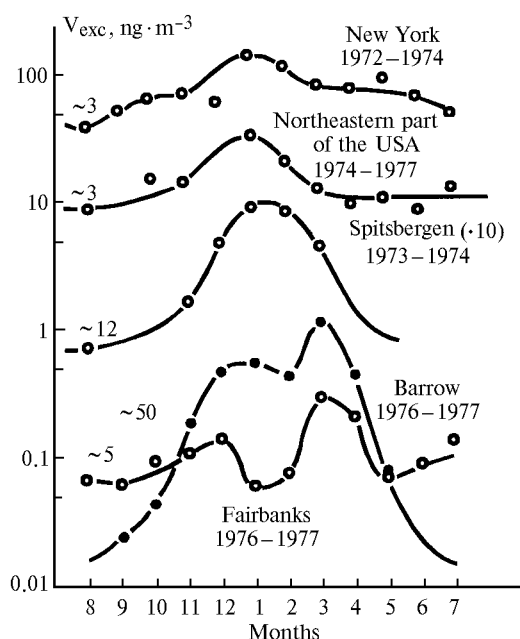


Fig. 13. Seasonal variability of excess (over the calculated concentration of the terrigenous component) vanadium, which is the indicator of the pollution transport in the Arctic in comparison with the northeastern part of the USA and New York.¹⁶⁵

These and other data indicate that every industrial center has its specific spectrum of pollution. On addition of these pollutants to the global silicate matter

and aerosol organic matter they mark them, thus being important tracers of air masses. Thus, the history of the aerosols origination and life in the atmosphere can be reconstructed from their composition.

We have succeeded in reconstruction of the air mass paths from the sites of aerosol formation to the sites of their complete or partial fallout using the combined analysis of the aerosol matter and the collection of meteorological maps through plotting the back trajectories of air masses. For long periods (seasons, years) these paths of sediment transport in the atmosphere can be reconstructed from the analysis of back trajectories; for even longer periods of time – from comparison with the data on aerosols, snow, glaciers, suspensions, crysols, and bottom sediments of lakes and seas. This line of "sedimentation and geochemical meteorology" is still in progress.

Such quantitative characteristics of the processes of aerosol transport and fallout as vertical flows of aerosol, mineral, chemical elements, and pollutants, are very important.

2.6. Flows of aerosols and the attendant pollutants

The vertical flows of the aerosol deposited on the sea or land surface can be estimated from the data on the aerosol content and size distribution. As was shown, away from the coastal areas the aerosol particle size distribution varies only slightly (see Fig. 4), what simplifies estimation of vertical flows. The above data on mineral and chemical composition and pollution of the aerosol matter can be recalculated both into the values for vertical flows of aerosol as a whole and for various minerals and chemical elements (pollutants).

This method involving some assumptions gives only rough values of the flows. When necessary, it can be supplemented and checked by direct determination of the flows by measuring the snow depth (estimation of flows for the period of snow cover), ice, and bottom sediments, including the measurements with the use of

various precipitation gauges, specialized water pools, etc. Besides, in recent years the methods of the isotopic analysis of flows (^{210}Pb , ^7Be , and other cosmogenic isotopes) receive increasing development.

The knowledge of the flows (not only the amount per unit air volume) gives the values specific for a given place and given synoptic situation, what is especially important when studying the regime of pollution fallout, and this apparently will be the basis for future ecological studies and monitoring.

The flows of the aeolian material to the sea surface in the Russian Arctic due to dry sedimentation were calculated by us on the assumption that the sedimentation rate of insoluble coarse ($> 1\ \mu\text{m}$) particles, which form the most, in mass, portion of aerosols (see above), is equal to $2\ \text{cm/s}$ (Ref. 166). The flows for the Arctic make up from 0.03 to $1.68\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ (on the average $0.39\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ or $142\ \text{mg}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$, at the standard deviation of $0.34\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$).

We have calculated the flow of insoluble aerosols a year assuming that the dry sedimentation is $1/4$ of the total value.⁵ We extend our summer data on the aerosol content to the whole year, taking into account that the aerosol content in summer is several times lower than in winter and spring,^{9,117,125,149} but the amount of aerosol sediments in summer is three to four times larger than in spring.^{2,167} The flow of the aeolian suspension calculated under these assumptions is equal to $570\ \text{mg}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$ ($1.56\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$).

The flow of the aeolian material to the drifting ice surface in the Arctic was determined independently based on the mean content of suspension in the fresh snow equal to $2.19\ \text{mg/l}$ (the mean from Refs. 34, 52, and 168–171, $n = 87$ cases) and the annually mean amount of atmospheric precipitation in the Arctic Ocean equal to $285\ \text{mm}$ (Ref. 172). These direct methods gave the value of $624\ \text{mg}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$ ($1.71\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$).

The value for the aeolian material flow measured in 1994 at the station Ni-Alesund on Spitsbergen by one more independent method (with a precipitation gauge) was $212\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ ($0.58\ \text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) (Ref. 53).

The average, over the three methods, vertical flow of aerosols in the Arctic comprises about $300\ \text{mg}/\text{m}^2/\text{year}$, what is far higher than formerly accepted. Of course, this value calls for further refinement.

2.7. Contribution of aerosol to formation of the Arctic nature

The presented data show that the contribution of aerosols to formation of the Arctic nature is more significant and diversified than it was considered earlier. First of all, this is the case of the aerosol material total amount and its seasonal distribution. According to different estimates, the total income of aerosol (insoluble part) to the Arctic Ocean (area of $13.1\ \text{mln. km}^2$) varies from 2.8 (measurements with a precipitation gauge on Spitsbergen⁵³) to 7.5 – $8.2\ \text{mln. ton}$ from our data and the estimates of the suspension content in the snow cover (Table 12). This is much less than the delivery of the river material to the Arctic (about $245\ \text{mln. ton}/\text{year}$) (Refs. 4 and 36). However, it should be kept in mind that the main portion of river suspension is deposited in marginal filters of the river/sea interface.^{173,174} Only about 7% passes through the filter, and only 5% reaches the area of the continental slope, that is, only $12\ \text{mln. ton}$ suspension passes through the filter. Contribution of the aeolian material to formation of the geochemical and mineral composition of the water suspension and crysol of arctic sea ice and bottom sediment is roughly equal to contribution of the suspended matter of rivers and sea ice material.¹⁷⁵ The income of organic matter from the atmosphere is also of great significance.

The study of the mineral and chemical composition of the arctic aerosols allowed us to divide them into three main natural groups:

1. The elements connected with the organic matter of plants (C, S, H, N, P, etc.).
2. The Earth's crust elements with the ratios typical of the Earth's crust matrix (Al, Si, Fe, Ca, Mg, Ni, etc.).
3. The elements connected with the sea salt and the ratios typical of the sea water matrix (Na, Cl, Br, S, Ca, Mg, etc.).

Commonly, aerosols represent mixtures of these three natural groups of elements in proportions varying in space and time. The group of anthropogenic elements (pollutants) occupies a specific place. Their content ($\text{EC} > 10$) nowhere in the Arctic achieves dangerous values. They serve markers of far and very far transport of aerosols from industrial regions of the temperate zone.

Table 12. Delivery of suspension to the Arctic Ocean

Method of delivery	Methods of analysis	Flows from the atmosphere, $\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	Delivery, $\text{mln. ton}\cdot\text{year}^{-1}$	Source
Aeolian transport	Netting	1.56	7.5	This work
	Suspension content in snow cover	1.71	8.2	Refs. 34, 137, 168–171
	Precipitation gauge	0.58	2.8	Ref. 53
River sink (before the marginal filter)	–	–	245	Refs. 4, 36
Ice transport from the Laptev Sea	–	–	4	Ref. 138

At very weak anthropogenic impact, the global aerosol background in the Arctic is well-defined (see Fig. 1). It is largely determined by aluminosilicates (the Earth's crust matrix), which are complemented in summer by sea salt and plant residues from the land. The global aerosol has sufficiently constant size distribution and mineral composition.

The aerosol delivery of biogenic elements (P, N, S, H), iron, and some other elements needed for life is of great importance for ecosystems of the Arctic, because their income with river sink is insignificant there. Aerosols regulate the primary production and biogeochemical processes both in the arctic seas and land. (*Azotobacter* does not develop under conditions of permafrost, what results in a deficit of nitrogen.) For example, nutrition of lichens is there rootless, i.e., exclusively atmospheric.

All the above-said leads to the conclusion that the actual role of aerosols in the Arctic was underestimated and new investigations employing non-standard approaches are needed.

Conclusions

1. The representative material on aerosols of the over-sea atmospheric layer has been collected in ten marine cruises to the Arctic and supplemented with measurements on the arctic islands.

2. The use of several complementary methods in the aerosol studies and the data on snow, on the ice surface, and sea ice has allowed us to estimate the amount and flows of the aerosol material in the Arctic up to the North Pole, its size distribution and mineral and chemical composition, and to identify the types of pollutants and regions of their origination. Numerous markers of the aerosol material were revealed and applied to determination of the areas of aerosol carry-over from the surface, trajectories of transport, and sites of deposition (deponents).

3. The distinguishing feature of the aeolian material in the Arctic is that its main delivery and unloading occur not in summer, but in winter, when the surface of feeding provinces (forest and tundra) is ice-bound and covered with snow. Aerosols of far and very far origin are of the main importance (especially, in winter), what is indicated by numerous markers.

4. In the Arctic seas and in the Central Arctic the aerosol unloads on the ice surface with snow most of a year (300–360 days), but not on the sea surface as in other zones. The zone of pack ice is characterized by round-the-year accumulation of the aeolian material for 3–15 years.

5. Aerosol unloading in the zone of pack ice happens at the places of ice melting – in the northern part of the Greenland Sea. Thus, the Arctic is a giant natural accumulator of the aeolian material and attendant pollutants, which first enter into ice, i.e., are transformed into cryosols, and then, many thousands kilometers far from the places of their fallout, into water.

6. The balance calculations based on our data show that the contribution of aerosols to formation of the sedimentary material in the Arctic is close to the contribution of the river sediments beyond the marginal filters of rivers (earlier the aeolian material in the Arctic was ignored). For many elements (Pb, Sb, Se, V, etc.) the aerosol source is the primary one.

7. The aeolian material plays an important role in delivery of organic matter and biogenic elements to the Arctic seas characterized by the deficit of biogens. According to our data, the aeolian material washed out by snow and rain provides the most part of elements needed for formation of the primary production of the arctic plankton.

8. Based on geochemical studies and analysis of pollutants, we have succeeded in determining the main local pollution sources (Norilsk, Kola Peninsula, oil flares of the Western Siberia). By the relations of elements, we have determined the spectrum of pollutants for each source (in the largest industrial regions) and followed the particular trajectories of the air masses passage in the Arctic, places of their unloading (deponents), and the seasonal behavior of the processes.

9. Our experience shows that along with the thorough study of the aeolian material in the atmosphere, the systematic studies of lithology and geochemistry of snow cover, which lives in the Central Arctic for 9–10 months, are necessary, since it is a natural collector of both dissolved and suspended aeolian material. The corresponding studies of lithology and geochemistry of the sea ice, on which surface the aeolian material is first deposited and then concentrated, are necessary as well. Thus, the study of the atmospheric matter acquires a systematic and multidisciplinary character, what sharply increases the possibilities of interpretation.

Now we are only at the first stage of these studies, and further thorough and long joint work is needed.

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