

## Heavy Metals in Atmospheric Air in the Kara Sea Water Area in September–October 2011

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**Abstract**—The article presents the results of measurements of Pb, Cd, Cu, Zn, Ni, Co Cr, As, and Fe concentrations in atmospheric aerosols in the surface layer over the Kara Sea. The data were collected on 12 segments of a cruise of the R/V *Akademik Mstislav Keldysh* in September–October 2011. The trace element contents in atmospheric aerosol samples were determined by atomic absorption spectrometry with electrical and thermal atomization and automatic background correction. The dataset was analyzed in relation to the direction in which air masses moved into the study area; the direction was determined from the reverse motion trajectories of air particles. It is demonstrated that concentrations of pollutants increase in air masses coming off the continent and decrease in air masses coming off the ocean. The enrichment of Arctic aerosols in trace elements is calculated.

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### INTRODUCTION

The modern growth rates of world production require geographical expansion in the search for new natural resource deposits. As a result, economic activity has been intensively encroaching on fragile natural zones, including, in particular, Russia's polar regions. An example is prospecting for raw hydrocarbon materials on the shelf of Russian Arctic seas, which will entail the creation of new infrastructure inside the Arctic Circle and increased transport load on the Northern Sea Route. Undoubtedly, such new emission sources will add to those already existing in the Arctic Circle, like the powerful heavy-metal-emitting nonferrous metallurgical centers of the Kola Peninsula and Norilsk and the less powerful Russian economic entities in the Svalbard and Novaya Zemlya archipelagos.

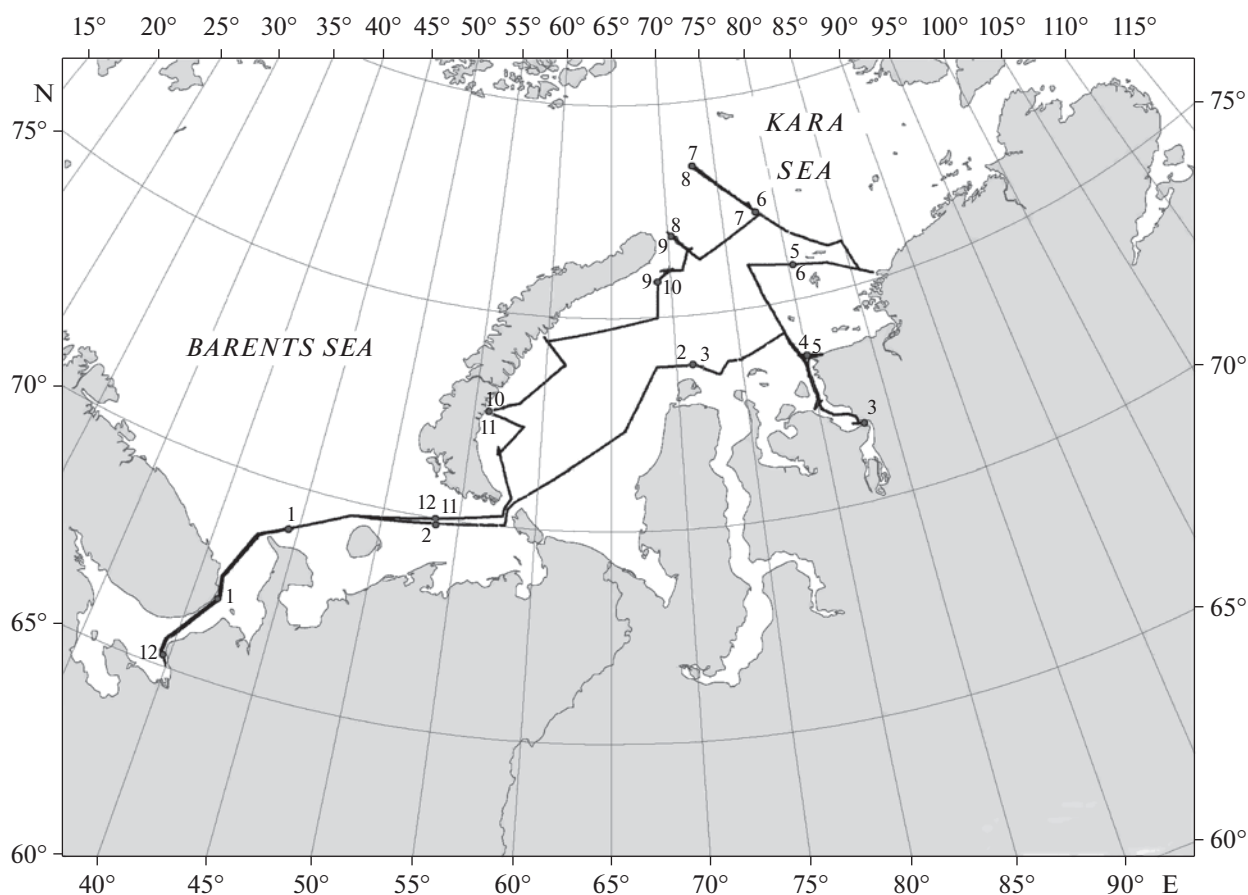
Simultaneously with the appearance of new sources emitting pollutants, the international adoption of a number of mandatory environmental protection measures since the 1990s has changed the priorities of emitters and affected global emission volumes [14, 15, 17]. However, anthropogenic emissions continue to prevail over terrigenous ones, e.g., Cd [18]; as a result, it is likely that some of the most toxic heavy metals will be transported to other natural environments, including Arctic marine ecosystems. Therefore, for Russian territory within the Arctic Circle, monitoring of the state of air pollution for a wide range

of substances remains urgent, including highly toxic heavy metals, primarily, Pd, Cd, Cu, and As. Among this group, Pb and Cd are included in the international European Monitoring and Evaluation program (EMER) for long-range transboundary air pollution [16].

The severity of the climate conditions and remoteness from economically developed territories hinder the creation of fixed regional air pollution monitoring stations on the Russian Arctic coast, the observational results of which could be used to assess the pollution dynamics for the Arctic's natural environment. Estimates of the modern (later than 2000) state of heavy metal air pollution (including Pb and Cd) continue to be based on sporadic measurements carried out in 2002–2010 in the Svalbard archipelago [7] on marine expeditions in 2001, 2002, and 2009 in open water areas of the Barents, White, and Norwegian seas from 32° to 50° E [4, 5, 13].

These studies in this sector of the Arctic show the following:

—atmospheric Pb, Cd, Cu, Zn, Ni, Co Cr, As, and Fe levels two or more orders of magnitude less than the maximum permissible daily average concentrations for the habitable zone and substantially less than the values characteristic of specially protected (from anthropogenic impact) Russian continental territories;



**Fig. 1.** Sampling sectors on route of R/V *Akademik Mstislav Keldysh* in September–October 2011 (numerals at beginning and end of sector correspond to its number).

—spatiotemporally, the values of concentrations vary within one order of magnitude;

—in comparison to the Barents and Norwegian seas, the air above the water area of the White Sea is more affected by heavy metal emission sources on the Kola Peninsula.

The aim of this study was to assess the current state of heavy metal pollution in the atmospheric surface layer in areas east of the Barents Sea using the same equipment and analysis methods as in [4, 5, 13] to ensure comparability of the results.

The article presents the results of measuring heavy metal contents in the surface layer from aboard the R/V *Akademik Mstislav Keldysh* on a Kara Sea cruise in September–October 2011.

## MATERIALS AND METHODS

Heavy metal contents in air above the sea surface were measured similarly to those in [4, 5, 13]. Samples were taken using an electric aspirator with a discharge rate of up to 60 m<sup>3</sup>/h, which sucked atmospheric air through Russian-made FPA-15 acetylcellulose filters [15] with an operational surface area of

320 cm<sup>2</sup>. This method of sampling atmospheric aerosols for subsequent determination of their chemical composition is used by Rosgidromet (Russian Hydrometeorological Service) background monitoring stations set up in biosphere reserves [10]. The aspirator was mounted on the upper deck of the *Keldysh* (the weather deck) 6 m above the water surface. The sampling period was from two to three days. The sample volume varied from 2500 to 2900 m<sup>3</sup>, which guaranteed in a working sample element concentrations more than an order of magnitude greater than in a blank run. The location of the aspirator prevented sample contamination from onboard emission sources. For the entire cruise, air samples were taken in situ at 12 sectors along the vessel's route (see Fig. 1).

Filter decomposition and measurements of the Pb, Cd, Cu, Ni, Zn, Co, Cr, As, and Fe concentrations were carried out by a method developed for low natural heavy metal concentrations in atmospheric air and used by the Rosgidromet integrated background monitoring system [11]. Measurements were performed on a KVANT-Z.ETA flameless atomic absorption spectrometer with the Zeeman effect for background correction.

**Table 1.** Characteristics of datasets of heavy metal concentrations in surface layer measured in water areas of Arctic seas in 2001–2011, ng/m<sup>3</sup>

Statistical parameters	Pb	Cd	Cu	Zn	Ni	Co	Cr	As	Fe
Kara Sea, September 2011									
Mean	0.54	0.012	23	7.7	3.2	0.05	1.1	0.07	41
Max.	3.0	0.1	70	43.0	4.6	0.2	2.5	0.18	121
Min.	0.18	0.003	7.3	2.2	1.6	0.01	0.41	0.03	19
Norwegian Sea, February–April 2010 [5]									
Mean	3.8	0.2	32	8.4	4.7	0.05	5.9		
Max.	8.8	0.75	147	39	8.3	0.14	14		
Min.	2.2	0.1	8.3	2.0	1.6	0.02	3.5		
Barents Sea, July 2001, 60° N									
Mean	0.13	0.023	3.8	24	0.3	0.19	0.31		40
Max.	0.27	0.032	5.2	47	0.85	0.36	0.45		82
Min.	0.06	0.01	0.34	12	0.12	0.064	0.14		15

Along the vessel's entire route, the following meteorological parameters were automatically recorded in the surface layer: wind direction and speed, atmospheric pressure, and air humidity. As demonstrated in [6], data on atmospheric processes are extremely important when interpreting the level of air pollution for geographical areas far from long-range emission sources. Therefore, the entire time the vessel was in the Kara Sea, the transport routes of air masses into the sampling areas were determined and the corresponding reverse motion trajectories of air particles were constructed.

Transport routes were constructed with a HYSPLIT hybrid Lagrange model for calculating reverse trajectories in the atmosphere [12] for up to five days prior to sampling, taking into account vertical movement along the route. Calculations used the meteorological parameter fields from two archives: data accumulated on a grid with a resolution of 1° × 1°, NCEP/NOASS Global Data Assimilation System (GDAS1, <http://ready.arl.noaa.gov/gdas1.php>) and reconstructed from reanalysis on a grid with a resolution of 2.5° × 2.5°, NCEP/NCAR Global Reanalysis Data (NCEP, [http://ready.arl.noaa.gov/gbl\\_reanalysis.php](http://ready.arl.noaa.gov/gbl_reanalysis.php)). For each hourly interval on a segment of the route, an individual reverse trajectory was calculated, which made it possible to obtain sets of air transfer trajectories for sampling periods of two to three days.

The calculation results show that a smaller spatial dispersion is more characteristic of trajectory sets obtained from the NCEP reanalysis data than for those obtained from the GDAS1 data. This is related to the low resolution of the initial grid of the meteorological parameter fields (2.5° versus 1°); however, on the whole, the calculation results from the data of different archives vary for no more than 30% of the trajectories; for the remaining sets, the difference in the calculated positions of air particle transfer projections

is insignificant. Our study used the analysis results for the GDAS1 dataset. Figure 2 shows examples of the calculated air mass transfer trajectories into the sampling areas.

As shown in Fig. 1, in the Barents Sea, part of the vessel's route passed close to the east coast of the Kola Peninsula, then along its southern areas (sectors 1 and 12) to the Kara Sea. A vast area in the Kara Sea was studied, bounded in the west by the island of Novaya Zemlya and in the south by the Arctic coast of Eurasia from 58° to 85° W. The northern part of the route was at a latitude of 78° N.

The air-sampling segments of the route were quite extensive. Sectors 1 and 12 and 3 and 4 were close to continental areas; sectors 10 and 11 were close to Novaya Zemlya; and sectors 5–7 were farthest from the continent.

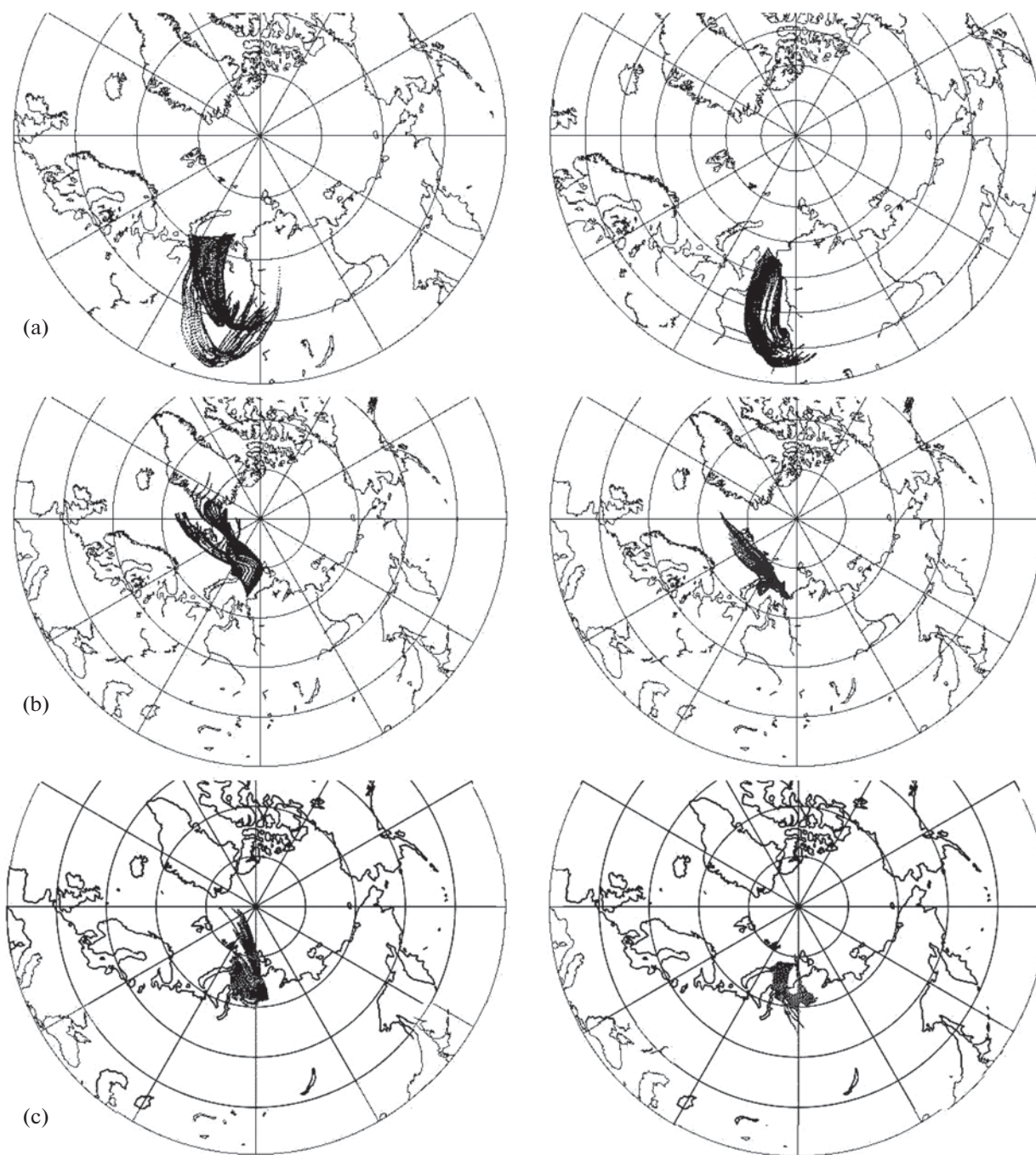
Atmospheric aerosol samples were collected as the vessel moved; therefore, the results of measuring their composition consisted of the spatiotemporally averaged characteristics for each sector of the route corresponding to the sampling time.

Thus, this measurement method in the water area of the Kara Sea made it possible to obtain the heavy metal content in atmospheric air in a remote marine area of the Russian Arctic with respect to the origins of air masses entering this region.

## RESULTS AND DISCUSSION

The data in Tables 1 and 2 on the concentrations of heavy metals in the surface layer obtained in the Kara Sea and similar data obtained in the Norwegian and Barents seas [3, 4, 14] show the following.

Heavy metal concentrations in the air over the Kara Sea water area, just like for those of the Norwegian and Barents seas, are substantially less than the



**Fig. 2.** Examples of calculated reverse trajectories in case of transfer from continent (a), from water area of North Atlantic and Arctic Ocean (b) and mixed (c).

values characteristic of specially protected continental territories [1] and two or more orders of magnitude less than Russia's maximum permissible hygienic norms for populated areas [2].

The characteristics of the Pb, Cd, Zn, and Fe concentrations near the 69th and 70th parallels in the Barents Sea (Table 1) and those in the atmosphere over the Kara Sea had the same order of magnitude. The

same can be said for the Cu, Ni, and Co concentrations (anthropogenic emission sources above the Arctic Circle) in the water areas of the Kara and Norwegian seas (Table 1).

For an equivalent domain of variation in the heavy metal concentrations in air for the considered Arctic seas, for the Kara Sea, the concentrations of heavy elements are just as spatiotemporally unstable as for the

**Table 2.** Mean values of heavy metal concentrations in surface layer at different latitudes of Kara Sea, ng/m<sup>3</sup>

North	Pb	Cd	Cu	Zn	Ni	Co	Cr	As	Fe
70°–75°	0.62	0.014	15.3	10.6	3.7	0.09	1.4	0.09	89
75°–80°	0.26	0.057	38	2.6	2.8	0.02	0.7	0.05	29

seas lying to the west. This is evidenced by the width of their variation range, bounded by the minimum and maximum values (Table 1) and for certain elements reaching several orders of magnitude (e.g., for Cd); this is also evident in the data of Table 2.

Detailed consideration of the results obtained in the Barents Sea (sectors 1 and 12 in Fig. 1) and the Kara Sea in 2011 show that in the dataset (on the whole, consisting of low concentrations), there are a group of high and a group of low element concentrations in the surface layer. To explain the formation conditions for concentrations of different levels, the heavy metal concentrations were analyzed in relation to the weather conditions resulting from the nature of the air mass in the measurement area.

Analysis of air trajectories into the area of the vessel's route (examples of which are shown in Fig. 2) and the meteorological parameters demonstrated that in the studied sea area, three starkly different types of weather conditions were observed, which formed under the action of the encroaching air masses; each type lasted 5–11 days.

The first type is movement of air masses off the continent to the ocean (Fig. 2a); in the surface layer, wind had the same direction, a speed of 10 m/s, and a temperature slowly dropping from 15 to 10°C; pressure also dropped slowly, and humidity varied weakly (characteristic of sectors 1–3).

The second type is air masses moving from open ocean areas (Fig. 2c): the wind direction changed to the north, its speed decreased to 4–5 m/s, the temperature dropped to 0°C, the pressure increased to the maximum then began to gradually drop, and humidity varied in a wider range (characteristic of sectors 5, 6, 10, and 11).

The third type is intrusion of a mixed (continental–marine, Fig. 2c) air mass: the wind direction varied from north to south at a speed of up to 15 m/s, the pressure continued to drop to 980 Pa, the temperature

rose to 3–5°C, and the humidity varied from 50 to 90% (characteristic of sectors 4, 7, 8, and 12).

Based on this analysis, air masses of different origin passed three times through the measurement area: off the continent (first type), from open ocean areas (second type), and mixed (third type), which generated the corresponding meteorological situation.

An increase in concentration for the entire group of heavy metals under consideration occurs in the measurement area when air masses off the continent intrude (Table 3). The concentrations increase by two to three times depending on the element—Cd, by more than an order of magnitude—than for those levels observed for oceanic and mixed air masses. The increased concentrations are retained when there is a continental air mass in the measurement area (up to five days or more).

The extent of the region of increased concentrations along the Russian Arctic coast is on the order of 1800 km, from the southern areas of the Barents Sea to the southeastern margin of the Kara Sea (sectors 1–3). Here, the spatial distribution of concentrations is non-uniform. The concentrations of the entire group of heavy metals, except for Zn and Co, is less than in sector 3 with respect to sector 1 (Table 4). The gradient of the decrease in concentrations to the east, depending on the element, is from 0.007 ng/m<sup>3</sup> per degree of longitude for Cd and up to 3.8 ng/m<sup>3</sup> per degree of latitude for Cu.

This indicates that air masses off the Eurasian continent have a greater influence on the state of pollution of the surface layer in southern areas of the Barents Sea (the western sector of Russian Arctic seas) than on southern areas of the Kara Sea.

The increased heavy metal concentrations in the continental air mass, in contrast to those that came from off the ocean, are apparently because it formed from anthropogenic metal aerosols, which is evidenced by an increased iron concentration (terrestrial marker), as well as from anthropogenic aerosols from continental sources. The closest anthropogenic sources of Cu, Ni, and Co emissions are the above-mentioned nonferrous metallurgical enterprises within the Arctic Circle.

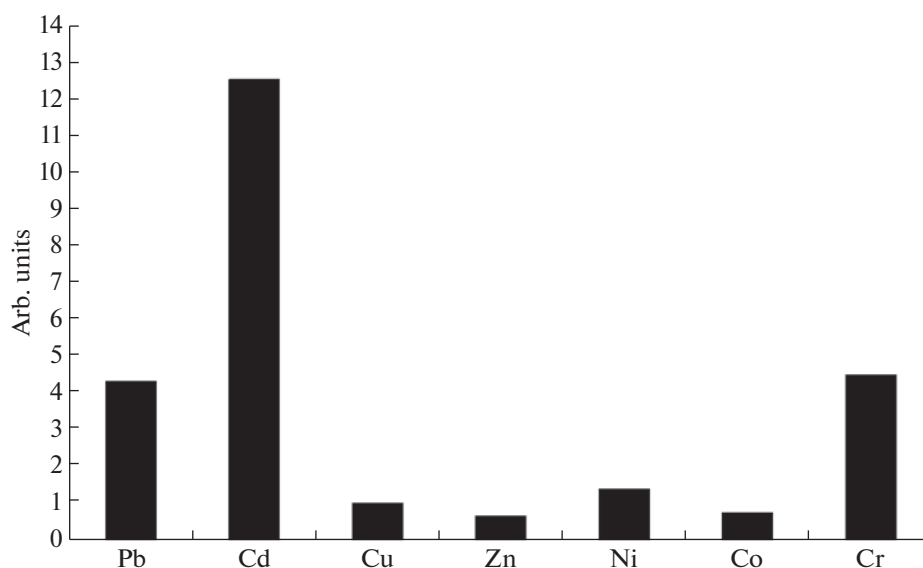
The anthropogenic component of heavy metals in aerosols in continental air masses is confirmed by data obtained in sector 10 near the Novaya Zemlya archi-

**Table 3.** Mean values of heavy metal concentrations in air of different Kara Sea areas for different air mass transfer directions, ng/m<sup>3</sup>

AM* transfer direction	Sector no.	Pb	Cd	Cu	Zn	Ni	Co	Cr	As	Fe
From continent (Fig. 2a)	1–3	3.1	0.18	82	23.5	6.7	0.13	1.6	0.37	95.0
From open ocean areas (Fig. 2b)	5, 6, 10, 11	0.3	0.006	21.5	4.2	2.6	0.04	0.86	0.07	40.0
Mixed (Fig. 2c)	4, 7, 8, 12	0.37	0.007	16.6	8.1	3.2	0.04	1.2	0.06	44.0

\*AM, air mass.





**Fig. 3.** Normalized heavy metal concentrations in air of Norwegian Sea water area in 2010 [5] with respect to concentrations in Kara Sea water area in 2011.

pelago (Fig. 1) for air masses that came from off the ocean. In this sector, the Fe concentration corresponded to values characteristic of continental air masses, while as before, the concentration of the remaining group of elements remained low.

The data for sectors 3 and 4 show which element concentrations decreased, how rapidly, and under what conditions (Table 4). The measurement interval in these sectors was two days. Within this time, the continental air mass was replaced by a mixed air mass, and in the surface layer, a decrease in the concentration of terrigenous Fe and such elements as Pb, Cd, and As occurred; the Cu, Zn, Ni, Co, and Cr concentrations changed but little.

In sectors 1 and 12 near the Kola Peninsula, measurements were carried out with a 20-day interval and for different characters of air masses in the measurement period. The results show that even in mixed air masses, the concentrations of the entire group of considered elements is lower than in continental air masses, while the Cu and Ni concentrations retain a level characteristic of the atmospheric surface layer for air masses coming off the ocean (Table 3).

Analysis of the reverse motion trajectories of air masses showed that in the measurement period, oce-

anic and mixed air masses prevailed in the Kara Sea. The concentrations of heavy metals in the marine atmosphere of the Norwegian Sea were measured under similar conditions. This made it possible to consider the spatial pattern of the concentration distribution in a wider sector of the Arctic (in contrast to the Barents–Kara Sea sector considered above). For this, we considered the mean concentrations of atmospheric elements for the Norwegian and Kara seas obtained during intrusion of air masses coming off the ocean, after which the mean values for the Norwegian Sea were normalized to those for the Kara Sea. The normalization results shown in Fig. 3 characterize the spatial distribution of element concentrations when continental (dirtier) air masses exert no influence. The character of the concentration field for each element can be judged by how much the normalized index differs from 1.

It is clear from Fig. 3 that even when oceanic air masses prevail in vast Arctic sea regions, the field of Pb, Cd, and Cr concentrations remains nonuniform; the anthropogenic sources of these elements are mainly located in densely populated areas. These elements express a stark trend of decreasing concentrations to the east. The spatial distribution of the Cu,

**Table 4.** Heavy metal concentrations in air of Kara Sea in sectors 1, 3, 4, and 12, ng/m<sup>3</sup>

AM transfer direction	Sector no.	Pb	Cd	Cu	Zn	Ni	Co	Cr	As	Fe
From continent	1	4.0	0.39	175	15.0	12.0	0.097	1.8	0.78	97.4
	3	3.0	0.1	22.0	43.0	3.6	0.096	1.0	0.18	68.0
Mixed	4	0.62	0.024	33.0	32.0	2.9	0.069	1.4	0.08	42.0
	12	1.8	0.0021	29.0	1.0	3.6	0.047	0.49	0.15	38.0

**Table 5.** Heavy-metal enrichment ratios for atmospheric aerosols with respect to mean composition of Earth's crust in Kara Sea air, 2011

AM transfer direction	Sector no.	Pb	Cd	Cu	Zn	Ni	Co	Cr	As
From continent	2	50	180	361	52	27	3.8	8.8	29
	3	116	490	288	319	38	3.2	7.4	66
	5	35	101	2562	33	85	2.7	12	78
From open ocean areas	6	34	105	2208	61	61	1.9	19	70
	10	21	65	118	77	46	2.9	9.8	25
	11	58	12	79	53	21	1.2	6.1	21
Mixed	4	39	190	701	385	50	3.8	17	48
	7	14	42	1330	64	60	3.0	16	41
	8	19	38	252	32	80	1.0	5.9	22
	12	124	17	681	13	69	2.9	6.5	99

Zn, Ni, and Co concentrations in the Arctic atmosphere can be considered nearly uniform. This is possibly the result of Cu, Ni, and Co emissions from copper and nickel smelting enterprises within the Russian Arctic Circle on the Kola Peninsula and in Norilsk, as well as Zn from the sea surface.

To reveal the degree of influence of anthropogenic emission sources for the considered group of metals on the composition of Arctic aerosols, we performed a joint analysis of the enrichment ratios ( $K$ ) of Arctic aerosols and the correlation coefficients of all considered elements with respect to iron for the air samples collected in the Kara Sea water area. We also took into account the character of the prevailing air masses.

The enrichment ratios were calculated with the widely used relation: a fraction whose numerator contains the ratio of element masses to the mass of iron in a sample and whose denominator contains the ratio of masses of the considered elements in the Earth's crust (after A.P. Vinogradov) [2]. The results are summarized in Table 5. The calculated correlation coefficients comprised the following decreasing series: Co, 0.85; Pb, 0.6; Ni, 0.55; Cd, 0.53; As, 0.49; Cr, 0.45; Cu, 0.30; Zn, 0.30. The critical significance level for the correlation coefficient at a 5% level was 0.58.

Based on the obtained  $K$  values ( $<10$ ) and significant positive correlation coefficient of 0.85 for Co and approaching the significance level for Cr, anthropogenic sources of Co and, to a lesser degree, Cr emissions play a very small role in the composition of Arctic aerosols. The concentration level of these elements measured in this study are apparently characteristic of the entire Arctic Basin.

When the level of  $K$  values for Pb, Cd, and Zn (up to 50 for Pb and up to 100 for Cd and Zn) are comparatively stable independent of the character of the prevailing air mass, the degree of atmospheric aerosol enrichment in these elements increases in the Kara Sea's coastal areas (Fig. 1, Table 5). The correlation coefficients for Pb and Cd with respect to Fe are close

to the significance level. This leads to the assumption that the anthropogenic component insignificantly exceeds the terrigenous component in the level of Pb and Cd concentrations (which on the whole are extremely low) in the atmospheric air of the Arctic. In this group of elements, Zn differs by a low correlation coefficient with respect to Fe, which speaks in favor of enrichment of Arctic aerosols in this element, not only due to anthropogenic and terrigenous sources, but also to its input to the surface layer by sea spray.

In contrast to Pb, Cd, and Zn, no increase in the enrichment ratios for Ni and As was revealed when the sectors of the vessel's route approached the coastline. The maximum  $K$  values, up to 85, were obtained under prevailing oceanic air masses. The  $K$  values for Ni and As for all the air over the Kara Sea are higher than that for Pb, but lower than for Cd. However, positive correlation coefficients with respect to Fe are close to the significance level. Such a combination of factors allows the conclusion that the level of influence of the anthropogenic component of Ni and As in the composition of Arctic aerosols is higher than for Pb and Cd.

Cu occupies a special place among the entire group of elements: the largest anthropogenic emission source is located in the Russian Arctic Circle. The  $K$  value for this element, obtained in sectors 5–7, for oceanic and mixed air masses reached values of 1330–2562. The correlation with Fe was low: 0.30.

Detailed analysis shows that a distinguishing feature of the samples collected in the indicated sectors is a high Cu content (two to four times higher than in the other samples) and an extremely low Fe content (remoteness from the continent). Such a combination of element masses in atmospheric aerosols in Arctic sea areas was found in the sector at the same latitude as Norilsk, which may be evidence that even for transfer of oceanic air masses, the presence of anthropogenic Cu is retained in amounts characteristic of continental background areas. From the results of measurements performed by the authors in 2016, the

concentrations of Cu in atmospheric air, e.g., in the Voronezh biosphere reserve, varied in the annual cycle within the same interval as in the surface layer in the Kara Sea.

The enrichment ratios calculated for aerosols collected in the surface layer in the Barents Sea in 2001 under prevailing mixed air masses [13] fall in the following ranges: Pb, 11–32; Cd, 117–365; Cu, 28–62; Zn, 153–607; Ni, 3–11; Co, 0.4–52; Cr, 1–8. The estimated ratios of Pb, Cd, Cu, Zn, and Ni enrichment of atmospheric aerosols collected in the surface layer of the Middle Caspian [8] vary from 10 to 40.

Comparison of the presented data with those in Table 5 show that the order of values for the ratios of enrichment of Arctic aerosols in the entire group of considered trace elements for the 10-year period hardly changed at all, with the exception of Cu. Atmospheric aerosols are enriched in copper, zinc, and nickel in Arctic sea areas to a larger extent than in the intracontinental marine area. The opposite pattern is observed for cadmium; for lead, the estimates virtually coincide.

### CONCLUSIONS

The results obtained in vast areas of the Kara Sea gave an idea of the modern concentration levels of the group of priority heavy metals in the surface layer of Arctic sea areas.

The heavy metal concentrations in the surface layer in the sector of Russian Arctic seas remains far less than the maximum permissible level for populated areas and less (with the exception of Cu) than in specially protected continental territories.

Heavy-metal-containing aerosols transported from the continent to northern latitudes temporarily increase their concentrations in marine areas adjacent to the continent, then as a result of changing meteorological conditions, they decrease to the lowest global levels, which are retained above the entire water area of the studied Arctic sea area.

The level of concentrations measured as air masses intrude from the Eurasian continent can be designated as increased with respect to the level of concentrations established above the marine water area for the arrival of oceanic and mixed air masses; it can be considered as representative, characterizing the intensity of heavy metal transfer from the continent to the Russian Arctic. Changing air masses facilitate a rapid reduction in concentrations. Estimates for the mean values of metal concentrations in the surface layer for oceanic and mixed air masses are close to each other.

On the whole, low concentrations of such elements as Pb, Cd, and Cr in the vast Arctic sector from 12° in the Norwegian Sea to 85° in the Kara Sea decrease to the east by two or more times depending on the element. The mean Cu, Zn, Ni, and Co concentrations remain at virtually the same level in the Arctic space.

Based on the enrichment ratios and correlation coefficients, the terrigenous component prevails for Co and Cr present in Arctic aerosols; for Pb and Cd, the influence of the anthropogenic and terrigenous components are the same; for Ni and As, the influence of the anthropogenic component is higher than for Pb and Cd; for Cu, the anthropogenic component predominates.

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