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Spatiotemporal Variations in Atmospheric Aerosol Characteristics over the Kara, Barents, Norwegian, and Greenland Seas (2018–2021 Expeditions)

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Abstract—We discuss the results from measurements of aerosol physicochemical characteristics in summer 2021 in the Greenland–Kara sector of the Arctic Ocean (the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*). The studied characteristics included: the aerosol optical depth of the atmosphere, the aerosol and black carbon concentrations, as well as the chemical composition of aerosol samples, i.e., the concentrations of the main elements, the concentrations of organic and elemental carbon, and the isotopic composition of carbon. For most aerosol characteristics, we noted lower average values as compared to the three previous expeditions. Taking into the consideration the data from previous expeditions, we estimated the differences in the aerosol and black carbon concentrations over the Kara, Barents, Norwegian, and Greenland Seas.

Keywords: atmosphere over the ocean, aerosol, black carbon, elemental and organic carbon, elemental and isotopic composition

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INTRODUCTION

Atmospheric aerosol plays an important role in the formation of climate and ecological states of the regions. In the last decade, increased attention has been devoted to the Arctic zone, most vulnerable to climate changes and anthropogenic impacts.

The aerosol characteristics (optical depth and particle concentrations) in the Russian sector of the Arctic were first measured by the staff of the Arctic and Antarctic Research Institute in 1970s–1980s from drifting stations "North Pole" and the Arctic islands [1–3]. At the second stage (since 1991), Academician A. P. Lisitsyn headed a more detailed study of physicochemical composition of aerosol onboard research vessels (RV) at Shirshov Institute of Oceanology, Russian Academy of Sciences [4–6]. In recent years, marine expeditions in the Arctic Ocean (AO) have become regular [7–13]. Moreover, aerosol studies in the Russian sector of the Arctic are carried out using data from observations at polar stations (Barentsburg, Tiksi, Cape Baranov) [14–16], from aircraft expeditions [17, 18], and from model calculations [19–22]. The totality of the data obtained makes it possible to replenish the deficient data on aerosol characteristics in the atmosphere over the AO.

In this work, we present measurements of aerosol characteristics from two new expeditions in the AO, i.e., in the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*. In addition, taking into consideration the data from previous expeditions, we carried out a comparative analysis of atmospheric aerosol characteristics over four Arctic seas: the Greenland, Norwegian, Barents, and Kara Seas.

1. CHARACTERIZATION OF EXPEDITION MEASUREMENTS

In 2021, we continued to study the atmospheric aerosol in marine expeditions, namely, in the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*. The aerosol optical and microphysical characteristics were measured using the set of instruments: portable sun photometer SPM, photoelectric particle counter AZ-10, and aethalometers АЕ-33 (MageeScientific) [23] and MDA-02 [24]. The methods of measurements and data processing were considered in previous works [11–13]. Therefore, we confine ourselves to a brief description of the characteristics analyzed and conditions of the expedition measurements.

The AZ-10 counter measurements were used to calculate: the number concentration of particles with radii 0.15–5 μ m (N_a); volumes of fine and coarse aerosol particles with radii smaller and larger than $0.5 \,\mu\text{m}$ (V_f and V_c). The aethalometers were used to measure the mass concentration of absorbing substance in the equivalent of elemental black carbon еBC [25]. In this work, we present only АЕ-33 aethalometer measurements.

The SPM photometer observations were used to determine: the aerosol optical depth (AOD) of the atmosphere $τ^a(λ)$; the Ångström exponent α; coarse and fine components of AOD (τ*^с* and τ*^f* at the wavelength 0.5 μm), as well as the atmospheric water content (*W*). The photometric observations were carried out in situations where the solar disk was not covered by clouds. This explains the discontinuity and shortage of some AOD observations.

The measuring instruments were placed at altitudes of about 15 (83rd cruise) and 12 m above sea level (84th cruise). The aerosol and black carbon concentrations were measured continuously and around the clock: every 11 min with AZ-10, and every minute with $AE-33$. The N_a and eBC measurements contain shortterm gaps and bursts (false measurements) due to the impacts from local sources of technogenic origin (polluted air from ventilator shafts and smoke from ship's funnel). Therefore, the measurements were filtered using a special algorithm [26]. This algorithm comprises the procedures of identifying defective data (gaps and bursts up to 3 h in duration) and their replacement by the average measurements taken before and after a given defect.

In addition to instrumental observations, the multichannel air blower was used to collect aerosol samples on filters and to subsequently determine the chemical composition. On average, aerosol sampling lasted for two days. Under unfavorable conditions (sea spray, smoke blown from ship's chimney), air pumping through the filters was terminated. The methods of laboratory analysis of the filters were presented in previous works [11–13]; therefore, we briefly clarify them. The elemental composition of aerosol samples (the K, Ca, Ti, Mn, Fe, Co, Ni, Cu, Zn, Br, Rb, Sr, Zr, Mo, and Pb concentrations) was determined by the method of Synchrotron radiation X-ray fluorescence (SXRF) analysis at Siberian Center of Synchrotron and Terahertz Radiation in the Institute of Nuclear Physics, Siberian Branch, Russian Academy of Sciences [27]. The content of elemental (ЕС) and organic (ОС) carbon was determined by the method of reaction gas chromatography [28]. The isotopic composition of carbon δ^{13} C in the aerosol composition was analyzed at the Tomsk regional center for collective use (Institute of Monitoring of Climatic and Ecological Systems, Siberian Branch, Russian Academy of Sciences) by the method of isotope-ratio mass spectrometry [29].

The route of the expeditions passed across the White, Barents, Kara, Norwegian, and Greenland Seas (Fig. 1). The aerosol characteristics were measured from June 18 to July 4 in the 83rd cruise of RV *Akademik Mstislav Keldysh* and from July 24 to August 23 in the 84th cruise. The volume of data from the two cruises totaled: 20 days of measurements of AOD; 45 days of measurements of the concentrations *N_a* and eBC; and 14 aerosol samples.

2. DISCUSSION OF RESULTS

2.1. Aerosol Optical and Microphysical Characteristics in the 83rd and 84th cruises of RV Akademik Mstislav Keldysh

In the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*, atmospheric aerosol was studied in a single season (summer) and in a single AO sector: from the Greenland Sea to the Kara Sea. Therefore, we consider the total dataset from two expeditions. In addition, there was no need to separately consider measurements for different seas. As was already noted before $[11-13]$, the variations in aerosol characteristics in individual expeditions $(1-2 \text{ months})$ were primarily caused by changes in air masses and weather conditions, and not by the spatial differences in aerosol over individual Arctic seas.

Figure 2 shows the variations in AOD and in concentrations N_a and eBC during two 2021 expeditions. The atmosphere in the period of the 83rd cruise stood out in very high transparency: the AOD in the

wavelength region 0.5 μ m ($\tau_{0.5}^a$) did not exceed 0.04. The AOD was characterized by higher values and larger variability range in the 84th cruise. In two epi- $(\tau_{0.5}^a)$

sodes $\tau_{0.5}^a$ reached 0.18: on August 14 in the north of the Greenland Sea and on August 22 in the south of the Barents Sea.

Trajectory analysis of air mass motion [30] and satellite maps of thermal anomalies (fire centers) [31] showed (Fig. 3a) that the high AOD values on August 13–14 were due to the transport at the altitude $h =$ 1500 m of eruption products from Krýsuvík volcanic system in Iceland (63.9 \degree N, 22.3 \degree W; August 11–17, 2021) [32]. The plume of outflow of volcanic aerosol was well manifested in the spatial AOD distribution plotted using MERRA-2 reanalysis data [33] (Fig. 3b). The AOD increase in this situation could also be influenced by the transport of polluted air from the territory of Great Britain and gas fields in the North

Fig. 1. Routes of the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh* (boxed numbers indicate AOD measurement numbers).

Sea $(h = 1000 \text{ m})$. We note that the volcanic and anthropogenic pollutants were transported at altitudes above 1000 m and, as such, had no effect on near-surface aerosol characteristics: air was supplied to the nearsurface layer from the AO territory $(h = 10 \text{ m})$.

A less certain explanation can be given to the increased AOD values on August 22–23, coupled with maximal concentrations of aerosol, black carbon ($N_a = 22.7$ cm⁻³, eBC = 2073 ng/m³), as well as ЕС (subsection 2.3). Air masses at all altitudes were carried from the water basin of the AO and north of the Canadian Arctic Archipelago. Thus, there is no evidence of long-range transport of any explicit pollutants. The ship in that period approached the continent in the region of the Rybachy Peninsula. The increased aerosol content seems to be due to local (breeze) transports of continental air to the coastal zone of the Barents Sea.

In addition to August 23, high *Nа* values were also observed on June 23 near Novaya Zemlya and on July 2–4 in the Pechora Sea. Short-term еВС maxima were recorded in the Dvina Bay of the White Sea (near Arkhangelsk) on June 20 and near the West Spitsbergen coasts on August 4. There are grounds to suppose that the concentrations are larger in these cases owing to proximity to continental sources of aerosol and heavier ship traffic.

Table 1 and Fig. 4 present the statistical characteristics of aerosol and the average spectral dependence of the AOD in the 83rd and 84th cruises calculated from hourly average values. A small amount of data obtained in the White Sea was not used in the calculations. On the whole, our characteristics correspond to the Arctic atmosphere and are consistent with data from our previous expeditions in Eurasian sector of the AO [11–13].

We turn attention to different relative variability ranges of aerosol characteristics. The near-surface concentrations N_a and eBC showed the largest variation coefficients of 153 and 385%. The AOD values show a moderate variation coefficient of 83%. The atmospheric water vapor content *W* and the Ångström exponent α (selectivity of spectral dependence of AOD) are the most stable: the variation coefficients are 25 and 38%, respectively.

Table 1. Statistical characteristics of aerosol in the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*: average and maximal (Max) values and standard deviations (SD)

Characteristic	Average \pm SD (Max)
τ_0^a 5	0.06 ± 0.05 (0.20)
α	1.02 ± 0.39 (2.45)
τ^f	0.041 ± 0.045 (0.16)
τ^c	0.019 ± 0.015 (0.06)
W , g/cm ²	0.48 ± 0.12 (0.80)
N_a , cm ⁻³	2.55 ± 3.90 (22.7)
V_f , μ m ³ /cm ³	0.26 ± 0.36 (1.93)
V_c , μ m ³ /cm ³	1.42 ± 3.09 (22.2)
eBC, $ng/m3$	$25.2 \pm 97(2073)$

Fig. 2. Variations in daily average AOD values at three wavelengths and in hourly average N_a and eBC values in the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*. Vertical lines border seas: BS stands for Barents Sea, KS for Kara Sea, NS for Norwegian Sea, and GS for Greenland Sea; numbers in the upper panel indicate the AOD measurement numbers.

2.2. Aerosol Optical and Microphysical Characteristics Based on Data from Five Expeditions

To determine the specific features of the spatial distribution of aerosol characteristics over the AO, we considered the total dataset of measurements in five 2018–2021 expeditions: the 71st [11], 80th [12], 83rd, and 84th cruises of RV *Akademik Mstislav Keldysh*, as well as "Transarctic-2019" [13]. We chose only these five expeditions, in contrast to [13], because it is necessary to compare the entire complex of aerosol physical (subsection 2.2) and chemical (subsection 2.3) characteristics obtained under similar (with respect to the regions and periods of measurements) conditions.

Table 2 presents the average values of aerosol optical and microphysical characteristics calculated for four Arctic seas. We note that the data (days of measurements) of aerosol characteristics over separate seas are not always enough to obtain representative statistical estimates. This is primarily true for the atmospheric AOD: from 3 to 12 days of measurements in

Fig. 3. (a) Back trajectories of air mass motion to the region of measurements (star) and fire centers (red circles); and (b) map of the spatial AOD distribution over the AO using MERRA-2 reanalysis data [33] for August 14, 2021.

different regions. Due to shortage of measurements, the difference in the average AOD values reaches a factor of five over separate seas. Estimates of the AOD characteristics are more reliable for the total dataset from AO Greenland–Kara sector (5° W– 100° E); they are presented in the last column of Table 2. The average AOD characteristics in this AO sector obtained during five expeditions and in the 83rd and 84th cruises (see Table 1 and Fig. 4) almost coincide.

Comparison of near-surface characteristics of aerosol over individual seas showed that the concentrations eBC, V_f , and V_c were the lowest in the atmo-

Fig. 4. Average wavelength dependences of AOD using data from the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh* and data from the five expeditions.

sphere of the Greenland Sea and maximal (except V_c) over the Barents Sea. The concentrations eBC and V_f are a little lower over the Norwegian Sea. This may be because the Greenland Sea is far from, while the Barents and Norwegian Seas are closer to, the sources of continental aerosol (the Scandinavian and Kola peninsulas). The high concentrations V_c over the Kara Sea can hardly be explained by natural causes: possibly, shortage of measurements might have an effect.

Comparison of data from the entire Greenland– Kara sector of the AO indicates that the average particle volumes $(V_f$ and V_c) and black carbon concentrations are a factor of 1.2–1.7 smaller in the 83rd and 84th cruises (Table 1) than in the five expeditions (Table 2).

2.3. Chemical Composition of Aerosol Samples in Greenland–Kara Sector of the AO

A large air volume should be pumped through filters for a long time (2–3 days) to determine the aerosol chemical composition. The samples collected in the 83rd and 84th cruises, as in the previous three expeditions, are still not sufficient for the comparative analysis of aerosol composition over separate seas. Therefore, the average characteristics of aerosol samples (Table 3) were calculated for the whole Greenland– Kara sector of the AO from measurements а) in the 83rd and 84th cruises and (b) in the five 2018–2021expeditions.

Figure 5 shows the variations in characteristics of aerosol samples (OC, EC, δ^{13} C) on the route of the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh*. From Fig. 5a it can be seen that the isotopic composition of carbon (δ^{13} C) in aerosol samples varied from −29.0 to −27.4‰. In most (8 out of 14) samples, we recorded a light isotopic composition (from -29.0 to -28.1% , which corresponds to the combustion of oil and gas products [34, 35]. In six samples, the δ^{13} C values were intermediate between light and heavy isotopic compositions: from -27.9 to -27.4% . These δ^{13} C values can be explained by mixed contribution from two carbon sources: combustion of oil products (light isotopic composition) and combustion of coal and wood burnt to give carbon with a heavier isotopic composition. The average isotopic compositions in the 83rd and 84th cruises (−28.2‰) barely differed from the average δ^{13} C value in the five expeditions in Greenland–Kara sector of the AO (−28.0‰).

The EC and OC concentrations in the period of the 83rd cruise (until July 5) did not exceed 61 and 380 ng/m3 , respectively (Fig. 5b). The carbon concentrations varied in a wider range in the 84th cruise. The largest organic carbon concentration (OC = 1685 ng/m^3) was recorded in a sample taken on August 6–9 near the Spitsbergen coasts. Like other aerosol characteristics (see subsection 2.2), the concentrations of ЕС were maximal (76.2 ng/m^3) on August 21–23 in the south of the Barents Sea in the approach to the Rybachy Peninsula. Despite these maxima, the average ЕС and ОС values in these two cruises turned out to be a factor of 1.9 smaller than, on average, in the five expeditions (Table 3).

The ЕС and еВС concentrations measured in the same periods (Fig. 5b) are largely consistent: the average difference between these characteristics is 19 ng/m^3 ,

Characteristic	Kara Sea	Barents Sea	Norwegian Sea	Greenland Sea	Total (four seas)
$\tau_{0.5}^a$	$0.018 \pm 0.007(7)$	0.094 ± 0.040 (12)	$0.054 \pm 0.013(9)$	0.077 ± 0.050 (3)	0.059 ± 0.05 (31)
τ^f	0.009 ± 0.003	0.062 ± 0.03	0.019 ± 0.015	0.053 ± 0.047	0.038 ± 0.04
τ^c	0.009 ± 0.007	0.030 ± 0.010	0.034 ± 0.003	0.023 ± 0.009	0.020 ± 0.013
α	1.03 ± 0.46	0.93 ± 0.2	0.41 ± 0.33	0.88 ± 0.28	0.94 ± 0.38
eBC, $ng/m3$	$26 \pm 27(22)$	$47 \pm 93(43)$	$37 \pm 55(33)$	18 ± 19.7 (17)	42 ± 101 (115)
V_f , μ m ³ /cm ³	0.45 ± 0.46 (15)	0.55 ± 0.55 (40)	0.48 ± 0.3 (30)	0.21 ± 0.25 (15)	0.35 ± 0.34 (100)
V_c , μ m ³ /cm ³	3.82 ± 3.98 (11)	1.62 ± 2.26 (37)	1.56 ± 1.7 (30)	1.07 ± 2.24 (15)	1.75 ± 2.92 (93)

Table 2. Average (\pm SD) values of aerosol characteristics over different seas based on data from five expeditions (the numbers of days of measurements are given in parentheses)

Fig. 5. Variations in (a) isotopic composition δ¹³C and (b) in concentrations OC and EC in the 83rd and 84th cruises of RV *Aka-demik Mstislav Keldysh* (horizontal segments correspond to the sampling periods).

and the cross-correlation coefficient is 0.6. At the same time, joint analysis of data from instrumental measurements and samplings in the 83rd and 84th cruises and from previous expeditions [11–13] revealed that the behaviors of separate aerosol characteristics sometimes show up opposite dynamics, which cannot be explained just by the difference in the characteristics themselves (microphysical and chemical compositions). The reason lies in the methodological features of measuring different characteristics: the method used, measurement mode, positions of air intake devices, etc. These instrumental measurements provide an option to identify and eliminate the cases of short-term technogenic ship impacts. We just can terminate air pumping through filters in the cases of the strongest (visually observed) technogenic impacts and only in daytime. That is, local sources onboard the ship can contribute to the collected aerosol samples, sometimes leading to suspicious results. To minimize the effect of this factor, we now manufacture a new air intake device [36] with a sensor of aerosol concentrations, which will automatically terminate air pumping through filters in the cases of technogenic impacts.

Table 4 presents the average aerosol elemental composition in the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh* in decreasing order of concentrations. Comparison with data from other expeditions in the Greenland–Kara sector of the AO showed multifold excess of the concentrations of elements in the 80th cruise of RV *Akademik Mstislav Keldysh*. The statistical calculations were modified to exclude data from the 80th cruise before the causes for large differences become clear.

From Table 4 (columns 2 and 3) it can be seen that the average concentrations of most elements in the

positions. It was interesting to estimate how the elemental composition of aerosol differs between the marine Arctic atmosphere and the continental midlatitude atmosphere. The comparison was performed using data obtained in a rural region of Siberia, i.e., Klyuchi settlement in Novosibirsk oblast (see column 4). Aero-

sol was sampled during summer 2013 [37]; and the concentrations of elements were determined again by the SXRF method [27]. The comparison of the two data types showed that the average concentrations of most elements are smaller over the Arctic seas than in the continental atmosphere. The strongest decrease (by a factor of 20–40) is characteristic for the Ti, Ca, Fe, Mn, Rb, and Zr concentrations. The average mass

83rd and 84th cruises are few tens of percent lower than in the four expeditions. The exceptions are very low (a factor of 2.8 lower) Ti concentrations and higher Ca and Ni concentrations. In their values, the concentrations line up in nearly identical sequences of their distributions in the two last cruises and in the four expeditions, except the medium group of elements (from Ti to V), showing somewhat different

Table 3. Average (\pm SD) values of $\delta^{13}C$, OC, and EC in aerosol samples collected in the Greenland–Kara sector of the AO (*n* is the number of aerosol samples)

Characteristic of aerosol samples	83rd and 84th cruises ($n = 14$)	Five expeditions $(n=39)$
$\delta^{13}C, \%o$	-28.20 ± 0.55	-28.00 ± 0.66
OC, $ng/m3$	496 ± 450	939 ± 1019
EC, $ng/m3$	27.9 ± 24.8	$54.4 + 60.4$

Element		83rd and 84th cruises ($n = 14$) Four expeditions except the 80th cruise ($n = 31$)	Klyuchi settlement ($n = 30$)
Ca	287 ± 285	261 ± 266	6989 ± 5059
K	156 ± 146	180 ± 176	2420 ± 1138
Fe	48.5 ± 45.9	76.1 ± 74.2	2182 ± 1380
Zn	14.1 ± 25.7	18.5 ± 22.5	28.3 ± 9.6
Br	11.4 ± 13.3	12.6 ± 12.4	2.2 ± 0.82
Cu	5.28 ± 5.79	5.91 ± 10.7	5.9 ± 2.9
Ti	5.04 ± 5.02	14.1 ± 15.2	614 ± 368
Ni	3.66 ± 9.47	2.63 ± 7.41	1.5 ± 1.3
Mn	2.42 ± 2.55	3.46 ± 3.86	87.6 ± 52.1
Cr	2.23 ± 2.29	2.39 ± 1.97	14.6 ± 10.1
Sr	1.52 ± 2.34	2.17 ± 2.01	15.6 ± 10.2
Pb	1.16 ± 1.19	1.26 ± 0.93	5.4 ± 2.9
V	1.13 ± 0.76	1.51 ± 0.86	23.3 ± 15.1
Co	0.49 ± 0.34	0.79 ± 0.68	10.5 ± 6.4
Rb	0.16 ± 0.12	0.16 ± 0.11	4.5 ± 3.0
Zr	0.15 ± 0.12	0.25 ± 0.29	6.3 ± 3.4
Se	0.13 ± 0.07	0.19 ± 0.12	0.15 ± 0.07
Mo	0.06 ± 0.04	0.09 ± 0.06	0.12 ± 0.08

Table 4. Average $(\pm SD)$ concentrations of elements $(ng/m³)$ in aerosol composition in the Greenland–Kara sector of the AO (*n* is the number of aerosol samples) and in Klyuchi settlement

concentrations of Zn, Cu, Ni, Se, and Mo are comparable at the Arctic and middle latitudes. An analogous feature was also noted by the authors of [38] who compared the elemental compositions of aerosol samples in the Arctic and southern parts of Siberia. Still another salient feature of aerosol composition in the Arctic atmosphere is a larger bromine content. The most probable Br sources are emissions from sea ice and snow [13, 39]. Owing to these features, the sequences of distributions of elements (decreasing from larger to smaller concentrations) differ between the Arctic atmosphere and the continental atmosphere, except first three elements, Ca, K, and Fe.

CONCLUSIONS

We present the measurements of the complex of atmospheric aerosol physicochemical characteristics on the route of the 83rd and 84th cruises of RV *Akademik Mstislav Keldysh* in the Greenland–Kara sector of the AO. The average aerosol characteristics over the two expeditions were: $τ_{0.5}^a = 0.06$ for $α = 1.02$; $N_a =$ 2.55 cm⁻³; V_f = 0.26 μm³/cm³; V_c = 1.42 μm³/cm³; eBC = 25.2 ng/m³; EC = 27.9 ng/m³; OC = 496 ng/m³; and $\delta^{13}C = -28.20\%$. Average values of most aerosol characteristics $(N_a, V_f, V_c, \text{eBC}, \text{EC}, \text{and OC}),$ obtained in the 83rd and 84th cruises, are a factor of 1.2–1.9 smaller than in five 2018–2021 expeditions. The average AOD and δ^{13} C values from the 83rd and 84th cruises coincide with those from the five expeditions. $\tau_{0.5}^a$

The concentrations of elements in aerosol samples collected in the 83rd and 84th cruises are few percent smaller than in the four expeditions in this same sector of the AO. The average concentrations of most elements in the aerosol composition are smaller over the Arctic seas than at midlatitudes. The largest (a factor of 20–40) difference in the concentrations is characteristic for Ti, Ca, Fe, Mn, Rb, and Zr. The salient feature of the elemental composition of aerosol in the Arctic atmosphere is the high (about 12 ng/m^3) Br content.

A joint analysis of data from sampling and instrumental measurements showed that certain aerosol characteristics sometimes show opposite dynamics of their behavior, caused not only by the difference in the characteristics themselves (microphysical and chemical compositions), but also by methodological features of their measurements. It is noted that the sampling technology should be perfected to eliminate or minimize the technogenic ship impact.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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