OPTICS OF CLUSTERS,
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Vertical Distribution of Alkanes in Atmospheric Aerosol in the Russian Arctic in September 2020

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Abstract—A combined experiment aimed at the study of the air composition over all the seas in the Russian Arctic was carried out in September 2020 with the Optik Tu-134 flying laboratory. The experiment included sampling the of atmospheric aerosol in the air layer from 200 to 9000 m above sea level and determining the concentration of saturated hydrocarbons (*n*-alkanes) in aerosol particles. Saturated hydrocarbon compounds in the range $C_9H_{20}-C_{27}H_{56}$ were found in the air of this sector of the Arctic. The main mass of this class of organic compounds is concentrated in the narrower range $C_{10}H_{22}-C_{20}H_{42}$. The concentration of *n*-alkanes in aerosol over all the seas was low (ranging within $9.3-12.6$ ng/m^3). The only exception was the Chukchi Sea, over which the concentration attained 37.7 ng/m^3.

INTRODUCTION

A more rapid increase in the air temperature in the Arctic in comparison with other regions of the planet is now at the focus of attention of the scientific community [1−3]. This can lead to very negative consequences for the nature on the Earth. Climate change in the Arctic region is already manifesting itself at many different levels in natural objects. Thus, it was shown in [4] that in the zone of continuous permafrost, the annual average soil temperature increased from 0.03 to 0.06°C/yr at a depth of 10−12 m. The base depth of permafrost achieved 8 m in discontinuous areas. In [5], it was revealed that an increase in the air temperature has led to changes in the hydrological cycle: the amount of precipitation, river discharge, and loss of continental ice. There are already a lot of such facts.

The warming of the Arctic climate can be already seen in changes in the composition of the air and, in particular, in the content of atmospheric aerosol [6−8]. The atmospheric aerosol was examined in numerous publications. However, as follows from the generalizing review [9], most of these publications deal with the nonorganic fraction of aerosol (ionic, mineral, or elemental composition) or black carbon.

As for the organic component of atmospheric aerosol, the information on it in the Arctic is scarce and scattered. This information mostly concerns the Western Hemisphere [10−20].

Sources of volatile organic compounds and their presence in the composition of aerosol particles were theoretically analyzed with the high-resolution GEOS-Chem CTM chemical transfer model [10]. The analysis revealed overestimation of biogenic sources of hydroxyacetone, methyl ethyl ketone, and glyoxal and underestimation of the power of sources of biogenic formic acid and production of peroxyacetic acid from biogenic and anthropogenic precursors. Atmospheric volatile organic compounds were measured using a time-of-flight mass spectrometer at Willum and Nord stations in northeastern Greenland from April to October 2018 [11]. The measured data showed that the content of acetonitrile and benzene peaked in August and September and reflected the air inflow from the continent, where biomass was burned. An increase in the sea surface contribution consisting of the inflow of carboxylic acids (formic and acetic acids and dimethyl sulfide (DMS)) was observed during the periods of snow and sea ice melting on the coast of southeastern and northeastern Greenland.

In September 2018, during the MOCCHA (Microbiology-Ocean-Cloud-Coupling in High Arctic) campaign, as a part of the 2018 Arctic Ocean expedition on icebreaker Oden (Sweden), the content of organic compounds in aerosol was measured using an iodide-adduct high-resolution time-of-flight chemical-ionization mass spectrometer. These measurements revealed the significant contribution of the sea

surface to formation of the near-surface aerosol. In particular, they confirmed that sulfur-containing compounds are products of oxidation of gaseous DMS released by phytoplankton and ice algae. The comparison of the time series of DMS oxidation products in the form of particles and the gas phase did not reveal a significant correlation, which indicates different lifetimes of the precursor and oxidation products in different phases.

The total amount of organic matter and its contribution to the total mass of aerosol was analyzed in [13, 14]. The total content of organic carbon in aerosol varied from 8 to 950 ng/m^3, with peaks in late summer, early fall, and late winter. The percentage of the water-soluble fraction was $57 \pm 11\%$ of the total mass on average throughout the campaign. In [15], it was found that sulfates and highly processed organic matter predominated in the chemical composition of the aerosol. The mass fractions of sulfate, organic matter, ammonium, and nitrate in the boundary layer of the North Atlantic were 69, 23, 7, and 1% and remained largely the same between seasons.

The vertical distribution of organic aerosol was studied in [16, 17]. In the lower troposphere, sulfates predominated in submicron aerosol (74% on average) [16]. Organic carbon, ammonium, and black carbon contributed 20, 4, and 1%, respectively. At higher altitudes, the ratio changed: the relative contents of organic carbon, ammonium, and BC were 42, 8 and 2%, respectively.

In [17], the field measurements were compared with some commonly used models. It was concluded that the tested models do not yet accurately describe the dynamics of the organic component in the atmosphere. The discrepancies between the simulated and measured data ranged within 30−60%. Some studies are aimed at identifying the components of aerosol particles, their origin, formation, and transformation in the Arctic atmosphere [18−20]. In particular, the average molecular distributions of the homologous series of *n*-alkanes, fatty acids, and *n*-alcohols in central Alaska are presented in [19] for warm and cold periods from June 2008 to June 2009. The average concentrations of all lipid components, including the concentration of n-alkanes, were significantly higher in the warm period than in the cold one. The carbon preference index (the molecular ratio reflecting the biological and anthropogenic origin of carbon) of alkanes varied from 2.2 to 21, and it was fivefold as high in the warm period as in the cold one thus indicating an increase in biogenic input from terrestrial higher plants in the warm period. It follows from this review that the studies of organic aerosol carried out in the foreign sector of the Arctic do not provide an unambiguous ideas of either its composition or spatial-temporal variability. In the Russian Arctic, the vertical distribution of organic carbon and its components has not been studied at all.

This paper partially fills the data gap for this region by presenting the results of measurement of the alkane concentrations in the boundary layer and free troposphere of the Arctic.

EXPERIMENTAL

The experiment was conducted in September 2020 onboard the Optik Tu-134 flying laboratory. The thorough description of the Optik flying laboratory can be found in [21]. The flights were carried out from September 4 to 17, 2020, over all the seas of the Russian sector of the Arctic: Barents, Kara, Laptev, East Siberian, Chukotka, and Bering, as well as over coastal territories.

To determine organic compounds in the aerosol, atmospheric air was sampled with the modified standard PPD-2M stream-pressure probe mounted instead of the window on the starboard side of the aircraft and directed towards the incoming flow, parallel to the fuselage axis. A sealed filter holder designed for membranes with a diameter of 50 mm (Sartorius PTFE 11803, pore size of 1.2 μm) was made of stainless steel and equipped with quick-release vacuum connections at the input and output. A rotary vane pump (Elmo Rietschle Gardner Denver V-VTN 26) with a gas meter (Shinagawa 2C-M) provided pumping and metering (in standardized liters) up to 25 m^3 of air per hour. Pneumatic communications located upstream of the filter holder are made of a Teflon tube with an inner diameter of 8 mm, while those located downstream the filters are made of a vacuum hose of the same cross section. The pump is powered by an inverter with a maximum power of 4.5 kW connected to a separate 27 V onboard power bus with a maximum possible current consumption of 100 A. For 20-minute operation, the setup for sampling the organic component allows 1 to 8 m^3 of air to be passed through the filter element. With sensitivity of 50 pg, the GCMS (gas chromatography-mass spectrometry) method allows recording alkanes at a concentration of 0.1 ng/m^3 in the atmospheric aerosol.

Because of the low concentration of organic aerosol in the Arctic region, we had to pump through large air volumes. The flight profile often changed at relatively short intervals. Therefore, to obtain representative aerosol samples for analysis, the sampling was conducted by the following scheme: in the layer from 200 to 2000 m over the sea surface and from 3000 to 9000 m in the free troposphere. Over the coastal areas, aerosol was sampled during flights from one airport to another, in the layer from 3000 to 9000 m. This procedure was performed over all the seas under study and during all flight. A diagram of one of the flights is shown in Fig. 1.

The flight over the Laptev Sea was carried out on September 9 (Fig. 1). After takeoff in Tiksi, the aircraft climbed up to 9200 m. The climb was followed

by 10-minute horizontal flight in the upper troposphere. Then, the aircraft successively descended to 5200 and 200 m in the north-north direction toward the New Siberian Islands and was horizontally flying for 10 min at each altitude level. At the lower level, the flight trajectory passed over the Sannikov Strait, southeast of Kotelny Island. It was followed by a climb to 9000 m, a turn to the western tack over Bennett Island, and a 10-min horizontal flight in the upper troposphere. Then, the aircraft again successively descended to 5200 and 200 m and performed 10-min horizontal flights at each level. At the lower level, the flight trajectory passed over the Laptev Sea northwest of Belkovsky Island. Further, the aircraft again climbed to 9000 m and was horizontally flying for 10-min in the upper troposphere. During the descent before landing in Tiksi, the aircraft successively reached the altitudes 5000, 500, and 1000 m and performed 10-min horizontal flights at each.

For each sea under study, aerosol was sampled on two filters: one in the layer from 200 to 2000 m (atmospheric boundary layer) and another in the layer from 3000 to 9000 m (free troposphere). Since the flight profile was variable, the filters were alternately switched depending on the flight altitude. Thus, the filters collected samples from the entire atmospheric layer over each sea, and they provide the average alkane concentration over the boundary layer and the free troposphere. The same is true for the flights between airports. Thus, for example, the sample taken on the Tiksi–Anadyr route represents the average over the entire route. Only this technique of "bulk" sampling has allowed us to collect samples sufficient for obtaining a well-identifiable quantitative picture for the entire spectrum of alkanes.

In total, 19 samples were collected, six of which corresponded to the lower level. The sample parameters are given below in Table 1.

The procedure of sample preparation and analysis of alkanes consisted of single extraction of a sample with chromatographic acetone (1.5 ml) in an ultrasonic bath and concentration of the extract on a rotary evaporator to 15 μL, followed by the analysis at a gas chromatography-mass spectrometer (50–250°C at a rate of 5°C/min, isotherm at final temperature of 45 min, without dilution, DB-1MS column). The analysis was conducted with the use of an Agilent 6890N chromatograph and an Agilent 5973N mass spectrometer. Hydrocarbons were identified with NIST 14 library databases of mass spectra, as well as by comparing the retention times of reference compounds in calibration mixtures, i.e., analytical standards for n-alkanes C_8–C_{40} (Fluka: Alkane standard solution C_8–C_{20}, Alkane standard solution C_{21}–C_{40}). A more detailed description of the procedure of sampling, sample preparation, and analysis can be found in [22].
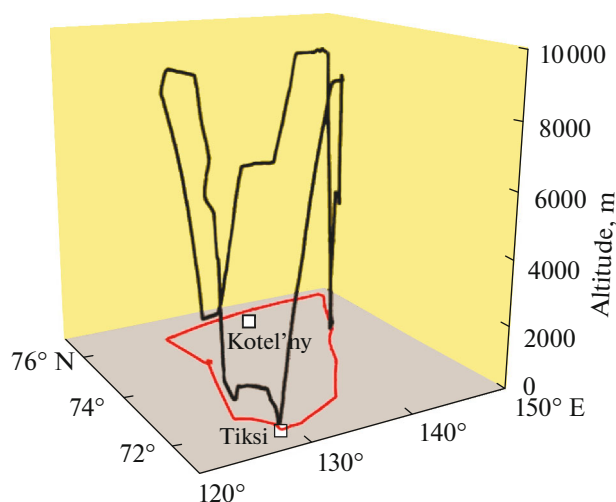

Fig. 1. Diagram of the flight with air sampling over the Laptev Sea.

RESULTS AND DISCUSSION

First, let us dwell on the total content of organic alkanes in the aerosol composition over sea areas. The data on the concentrations in the samples taken over the corresponding seas in the altitude layer 200–2000 m are given in Table 2.

It follows from Table 2 that the alkane concentration over all the seas ranged within 9.3–12.6 ng/m³. The exception is the Chukchi Sea, over which the concentration attained 37.7 ng/m³.

High concentrations of pollutants over the Arctic are usually associated with the transfer of pollutants to the Arctic region from the continent [23–26]. This is judged from the back trajectory analysis. This technique is inapplicable in our case, since the area under study covers hundreds of square kilometers. The synoptic analysis of the direction of air mass transfer generally indicates that air masses came to the Barents, Kara, and Laptev Seas from the continental Europe, to the East Siberian Sea, from the eastern sector of the Arctic Ocean, and to Bering Sea, from the Pacific Ocean. Only when the flights were carried out over the Chukchi Sea, the findings indicated the air mass transfer from Alaska. This is clearly seen from Fig. 2, which shows the synoptic map for the period under study.

We have no measurement data for the Alaska region, but the earlier published results indicate that this region could well be a source of high alkane concentrations. As was shown in [27], the total amount of the organic component in aerosol generated at the American continent can be up to 2 μg/m³. The content of the alkane fraction in Alaska can attain 50 ng/m³ in the warm period [19]. The damping property of the ocean is known, which consists in lagging the maximum and minimum of the annual variation in the air temperature relative to the annual variation in insola-
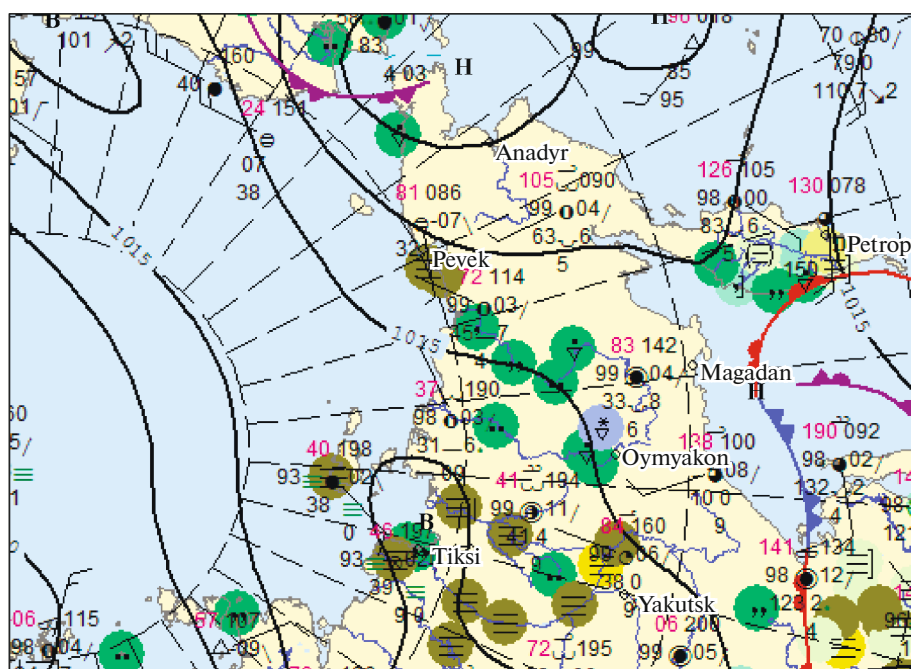
Fig. 2. Synoptic weather map on September 16, 2020, 00:00 UTC for the region under study.

Table 1. Sampling site and altitude and air volume pumped

No.	Route	Altitude, m	Air volume, m^3
1	Tolmachevo−Arkhangelsk	>3000	18.8
2	Barents Sea	3000−9000	6.7
3	Barents Sea	200−2000	4.2
4	Arkhangelsk−Naryan-Mar	>3000	5.6
5	Kara Sea	3000−9000	8.5
6	Kara Sea	200−2000	4.1
7	Naryan-Mar−Sabetta	>3000	4.1
8	Sabetta−Tiksi	>3000	9.5
9	Laptev sea	3000−9000	8.4
10	Laptev sea	200−2000	6.4
11	Tiksi−Anadyr	>3000	10.7
12	Chukchi sea	3000−9000	7.2
13	Chukchi sea	200−2000	4.6
14	East-Siberian Sea	3000−9000	7.6
15	East-Siberian Sea	200−2000	4.5
16	Bering Sea	3000−9000	2.5
17	Bering Sea	200−2000	4.8
18	Anadyr−Yakutsk	10600	11.3
19	Yakutsk−Tomsk	9800	13.4

Table 2. Total concentration of n-alkanes (ng/m^3) over the seas of the Russian Arctic

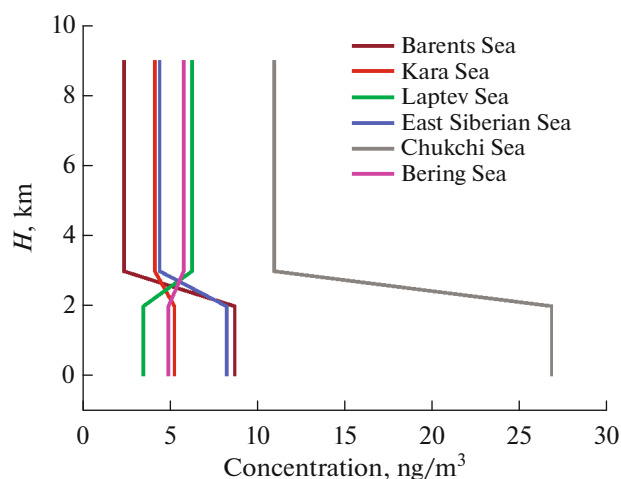| Sea	Barents	Kara	Laptev	East Siberian	Chukchi	Bering
Concentration	11.0	9.3	9.7	12.6	37.7	10.6

Fig. 3. Vertical distribution of *n*-alkanes over the Arctic seas.

tion [28]. Our measurements were carried out in the warm period, and the obtained alkane concentration is close to that recorded in Alaska, although, certainly, this is not a direct proof of the transfer from Alaska.

Our long-term measurements of saturated hydrocarbons over the continent have shown that their total concentration usually exponentially decreases with altitude [29]. However, there are some deviations from this pattern [30]. Figure 3 shows the vertical distribution of the aerosol under study over the Arctic seas.

It can be seen from Fig. 3 that the alkane concentrations vary within $3-8$ ng/m^3 in the atmospheric boundary layer and $2-6$ ng/m^3 in the free troposphere. The exception is again the Chukchi Sea, where the *n*-alkanes concentration was three times higher than over other seas in the boundary layer and two times higher in the free troposphere. These data are comparable with the results obtained for these saturated hydrocarbons in [19].

Considering the character of the vertical distribution of alkanes over the different Arctic seas, we can notice that the alkane concentration in atmospheric aerosol decreases with altitude over four seas and increases over two seas (Laptev and Barents seas). This vertical distribution is not an exception. The possible inflow of pollutants to the Arctic from the upper tropospheric layers was observed in [16, 17] and theoretically stated in [31].

The composition of organic components of aerosol particles in the layers from 0.2 to 2.0 km from $3.0-9.0$ km is shown in Fig. 4 on the logarithmic scale.

Alkanes from C$_9$ to C$_{27}$ are detected in the composition of aerosol particles over the Arctic seas (Fig. 4). This range is much narrower than that observed in the surface are layer in Arctic [18, 19] and far smaller than that observed over continental regions [32]. In view of the logarithmic scale, we can say that alkanes in aero-

sol particles are basically represented by C$_{10}$–C$_{22}$ compounds. Heavier compounds are detected in less than half of the cases, over some seas. The concentration of the compounds under study is slightly lower in the boundary layer and free atmosphere than in the surface air layer over the continental part and seas in the western Arctic [18, 19]. The ranges of light C$_n$H$_{2n+2}$ compounds in the boundary layer (Fig. 4a) and in the free troposphere (Fig. 4b) are nearly identical, although discrepancies are observed for heavier homologues with $n > 20$, which is probably due to the higher selectivity of their sources and the different origin of air masses in the layers under study.

A somewhat different distribution of organic compounds was observed over coastal regions (Fig. 5). Due to the peculiarities of sampling, Fig. 5 should be compared with Fig. 4b, only for the free troposphere. There were no measurements in the atmospheric boundary layer over land, since the aircraft quickly flies through the 0−2000-m layer, and it is impossible to accumulate a sufficient amount of matter for the reliable analysis.

It can be seen from Fig. 5 that heavy homologues C$_{23}$H$_{48}$–C$_{29}$H$_{60}$ are significant contributors to the composition of particles over coastal regions, especially western ones. This is likely due to differences in the vegetation in these areas.

According to the studies [33, 34], the presence of C$_9$H$_{20}$–C$_{14}$H$_{30}$ compounds in the composition of particles can be attributed to natural biological products: gases, alcohols, and acids emitted by vegetation. According to the data of [35, 36], C$_{15}$H$_{32}$–C$_{22}$H$_{46}$ are anthropogenic aerosols. They include lubricating oil and diesel fuel (C$_{19}$H$_{40}$) and car exhaust (C$_{21}$H$_{44}$ and C$_{22}$H$_{46}$). C$_{23}$H$_{48}$–C$_{29}$H$_{60}$ are mainly of natural biological origin [37, 38]. This separation is relatively conventional, since all these compounds can be present, for example, in petroleum products [39]. The separation is based on compounds prevailing in each mode.

Based on this classification and the data shown in Figs. 4 and 5, we can conclude that most of alkanes in aerosol over the Arctic seas and coastal regions are of natural biological origin. This conclusion could be verified using CPI (carbon preference index) suggested as early as in 1961 [37]. CPI is calculated by the formula, which includes compounds from C$_{24}$H$_{50}$ to C$_{34}$H$_{70}$ [41]. However, as can be seen from Figs. 4 and 5, there are very few or no such compounds in our samples. That is why CPI can hardly be applied for this assessment in our case.

In conclusion, let us dwell on the following aspect. The constructed back trajectories of air masses arrived to the regions under started on the continent in most cases, with the exception of the East Siberian Sea. Synoptic maps for different altitudes show the same. In this case, the alkane concentrations in aerosol over the coast should be higher or comparable to the con-
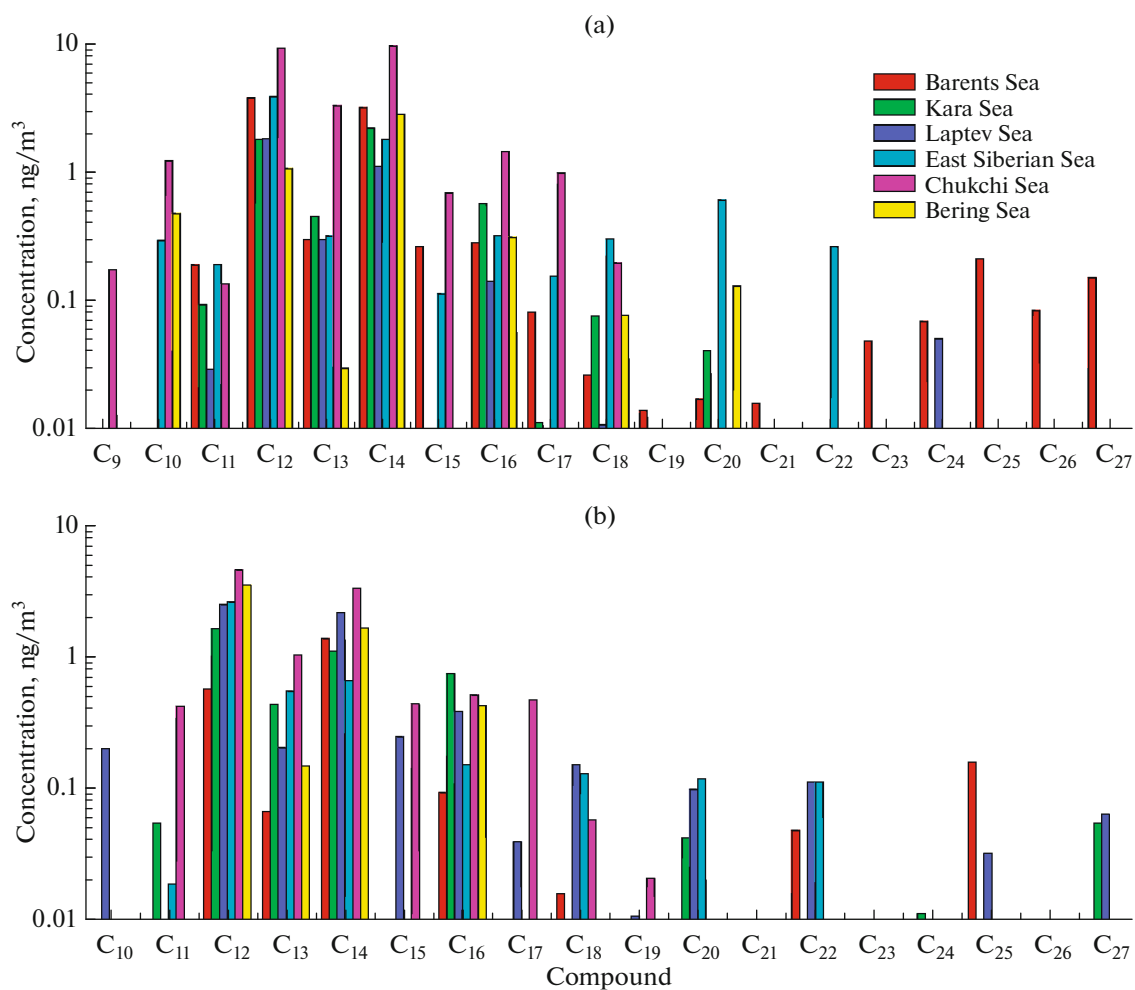
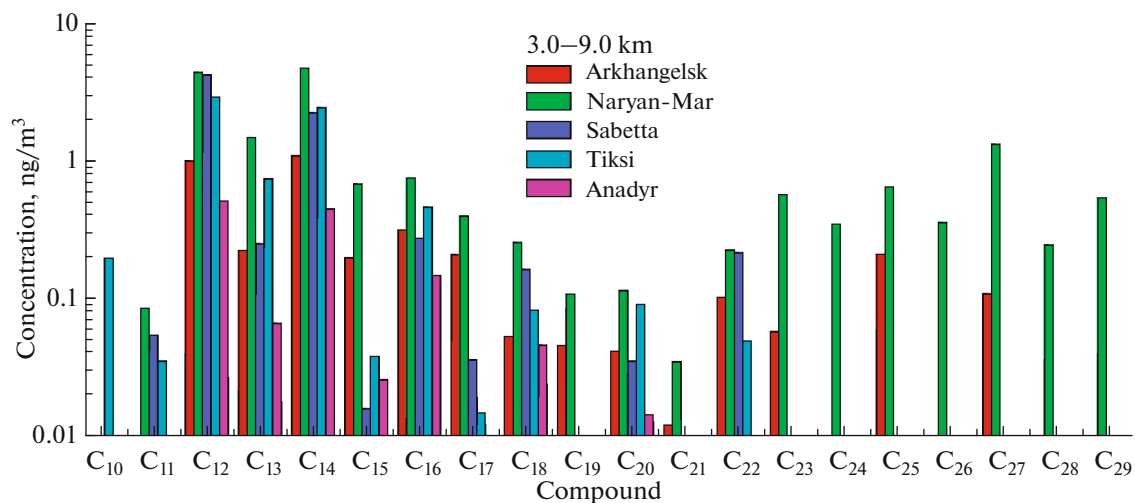**Fig. 4.** Alkanes over the Arctic seas in the layer (a) 0.2–2.0 and (b) 3.0–9.0 km.
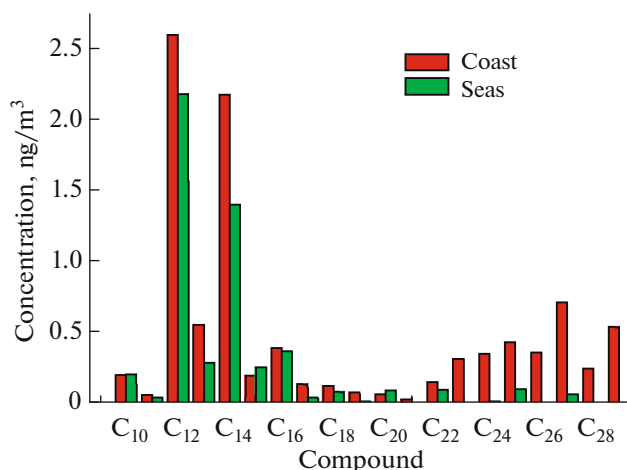

Fig. 5. *n*-alkanes over coastal Arctic regions.

Fig. 6. Composition of alkanes over the Arctic coast (6 samples) and seas (5 samples).

centrations recorded over the seas. For comparison, Fig. 6 shows the average concentrations over the coast and over the seas. The Chukchi Sea, as a special case, is excluded from the comparison.

The concentrations over the coast are obviously higher than or equal to the concentrations over the seas for almost all the compounds (Fig. 6). In our opinion, this indicates that organic compounds are indeed transferred to the Arctic water areas from the continent.

CONCLUSIONS

The study of the distribution of alkanes over the seas of the Russian Arctic has shown that their concentration is $9-12$ ng/m^3, which is comparable to the concentration of n-alkanes over adjacent foreign sectors of the Arctic.

The composition of aerosol particles persistently includes alkanes $C_9H_{20}-C_{19}H_{40}$; this range is much narrower than that observed over continental regions and adjacent territories.

The altitude distribution of the alkane content shows the downward trend over most of the seas, although over two seas (Laptev and Bering seas) the trend was upward, which is likely due to the long-range transfer of aerosol in the upper atmospheric layers.

The transport of alkanes with the aerosol from the continental Eurasia to the Arctic atmosphere is confirmed by the comparison of their concentrations over coastal and sea areas, as well as by the data of synoptic analysis.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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