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MARINE CHEMISTRY

The Concentration and Composition of Hydrocarbons in Water, Particulate Matter, and Bottom Sediments of the Kara Sea

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Abstract—Data are presented on the content of aliphatic and polycyclic aromatic hydrocarbons (AHC and PAH, respectively) in the interstitial waters and bottom sediments of the Kara Sea compared to the distribution of the particulate matter and organic carbon. It was found that the AHC concentrations within the water mass (16 μg/l on average) are mainly formed by natural processes. The AHC distribution represents the variability of the hydrological and sedimentation processes in different regions of the sea. The widest ranges of the concentrations were registered in the Ob Bay–Kara Sea section: in the water (10–310 μg/l for the AHC and 0.4–7.2 ng/l for the PAH) and in the surface layer of the bottom sediments (8–42 μg/l for the AHC and 9–94 ng/g for the PAH). The differentiation of the hydrocarbons (HC) in the different media follows the marginal filter's regularities; therefore, no oil and pyrogenic compounds are supplied to the open parts of the sea. In the sediment mass, the HC content is determined by the variations in the oxidative conditions in the sediment and its material's composition.

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INTRODUCTION

The studies of hydrocarbons (HC) in high-latitude regions are becoming especially topical in view of the exploration of oil and gas resources in the Arctic sea shelf. Because of this, the development of monitoring to determine the conditions in these aquatic areas, including the transboundary supply of pollutants, is becoming one of the main problems that must be solved to provide for their environmental safety. In the course of the monitoring, one must take into account that the anthropogenic HCs extend in the stable biogeochemical background existing in nature. Whereas these concentrations are zero for xenobiotics such as pesticides, the situation is more complicated for oil HCs. The phytoplankton annually produces 12 million tons of HCs, and, according to the resent data of the National Research Council (USA), the mass of the anthropogenic HCs supplied from all the sources amounts to 1.3 million tons per year on average (with a potential range from 0.47 to 8.4 million tons per year) [24]. The studies performed showed that natural (not caused by human activity) geochemical anomalies are permanently present in the marine environment [11]. Without analyzing the hydrocarbon concentrations, one cannot distinguish their anthropogenic constituent. Of special importance is the researching of the geochemical barrier zones where the processes of dispersion and concentrating of various compounds are the most intense (river–sea, atmosphere–water, water–particulate matter, etc.).

To study the processes of the supply and differentiation of the natural and anthropogenic HCs, as well as their potential transformations under different forms of migration, in the course of cruise 54 of R/V *Akademik Mstislav Keldysh* (September 2007), research of the AHC and the PAH in the water, the particulate matter, and the bottom sediments was performed. Special attention was paid to the Ob River–Kara Sea barrier zone.

The hydrological conditions in the Kara Sea, especially in its southwestern part, are determined by the riverine runoff, mainly by that of the Ob and Yenisei rivers. About 58 and 80% of the annual riverine supply fall in May–July and June–September, respectively. The development of industries and the construction of towns, dams, and power plants has become one of the main causes of the variability of the riverine flows [22]. The hydrocarbons supplied by the Ob and Yenisei rivers are spread over the vast expanses of the Arctic Basin. The particulate matter formed is enriched in various compounds and constitutes a sort of geochemical traps providing the accumulation of the riverine runoff components, including anthropogenic ones [7]. Therefore, the riverine matter is precipitated at the river–sea boundary, which determines the content of organic matter (OM) and of the HC within its composition and, hence, takes on the meaning of a global factor [5, 17].

The studies previously performed in the mouth areas of the Ob and Yenisei rivers (cruise 49 of the

R/V *Dmitry Mendeleev* in 1993) allowed one to create a model of the transformation and sedimentation of various compounds in the area of the river–sea geochemical boundary (a marginal filter) [7]. According to this model, the marginal filter consists of two key parts that are fundamentally different in their functions: the abiotic part adjacent to the river's mouth and the biotic part situated near the seaward side of the mixing zone [4]. In turn, the abiotic part consists of two zones: the gravitation zone, in which the sedimentation of the sandy–silty fractions proceeds, and the physicochemical zone, where the colloids and dissolved compounds are trapped (the zone of flocculation and coagulation). These zones are characterized by high water turbidity and hindered photosynthesis. After the precipitation of various compounds and the water's clarification, the phytoplankton development takes place and a biological zone of the assimilation and transformation of the mineral and organic dissolved substances appears ("a phytoplankton pump") [7]. At the zooplankton part of the filter, the filtration of the entire variety of suspended particles of different origin is seen.

METHODS OF THE STUDIES

The water was sampled with a bucket from the surface during the vessel's movement and when approaching the stations and with a plastic bottle sampler from the deep layers. The bottom sediments were collected using an Okean dredge, a Neimisto tube, and a large diameter tube (LDT). The particulate matter was separated by means of GF/F glass-fiber filters (0.7–1.2 μm). The hydrocarbons were extracted on board the vessel (immediately after the sampling) with chloroform from the water and with methylene chloride from the moist samples of the particulate matter and from the bottom sediments predryed at 40°C. The lipids (the total extractable fraction) and the HC, respectively, were determined before and after the column chromatography using silica gel. The AHC and PAH, respectively, were separated with hexane and a 3 : 2 hexane–benzene mixture. The concentration of the lipids and the AHC was determined by means of the IR technique using the 2930 cm^{-1} band with an IR 435 Shimadzu device (Japan) [9]. As a standard, a mixture of 37.5% isooctane, 37.5% hexadecane, and 25% benzene was used. To convert the AHC concentrations into those of the C_{org}, the parameter of 0.86 was used [10].

The content and composition of the PAH were determined using the HPLC technique using a Milikhrom A02 Ekonova devise (Russia) equipped with a Nucleosil–120–5–C_{18} column. As an eluent, a 75 : 25 mixture of acetonitrile and water was used in a gradient mode (up to 100% acetonitrile). The measurements were carried out at 254 nm. The identification was realized by the retention times of the individual polyarenes provided by the laboratory of the Environmental Protection Agency (USA). The procedure's sensitivity amounted to 0.4 ng with a peak due to anthracene. As a result, the following unsubstituted polyarenes were identified: phenanthrene (Ph), anthracene (A), fluoranthene (Fl), pyrene (Py), triphenilene (TPh), chrysene (Chr), perylene (Pl), benz(a)pyrene (Bp), benz(k)fluoranthene (BkFl), and 1,12-benzperylene (BPl).

The organic carbon in the bottom sediments was determined by dry combustion with an AH 7529 analyzer [8].

RESULTS AND DISCUSSION

In the waters of the open part of the Kara Sea, the range of the content variations in the surface layer amounted to 5.2–31.1 μg/l for the AHC_d (the dissolved form) and 3.2–26.8 μg/l for the AHC_p (the particulate form).

In the Kara Sea–Yamal Peninsula section, the AHC_d content varied from 5.2 to 12.3 μg/l (7.6 μg/l on average) with that of the AHC_p being from 3.2 to 19.2 μg/l (10.4 μg/l on average). The concentration variability for the AHC_p was a little wider than that for the AHC_d (σ of 0.6 and 0.4, respectively), which might be caused by the lability of the proper particulate matter. The Yamal current that arises near the Karskie Vorota Strait supplies the Barents Sea waters, which are evidently enriched in organic compounds (OC)—dissolved (DOC), particulate (POC), lipids, and HC—because the DOC concentrations at the seaward stations (2.17–3.15 mg of C/l) were also found to be higher than those near the Yamal Peninsula (1.59 mg of C/l; the data from Belyaev's report on the studies during cruise 54 of the R/V *Akademik Mstislav Keldysh*, vol. 2). This conforms as well to the former results obtained in the Kara Sea, where the primary production was evaluated as 170 and 25 mg of C/m^2 day in the western and eastern part of the sea, respectively [17]. However, the HC concentrations in the open waters and near the Yamal Peninsula are little different despite the increase of the fluorescence intensity and the chlorophyll in this direction [1]. At the same time, the maximum content at station 4960 in the open sea complies with the local increase of the particulate matter and the chlorophyll concentrations at the surface (up to 0.6 mg/l and 1.4 mg/m^3, respectively), which is probably caused by the phytoplankton development [1]. Evidently, the phytoplankton affects the AHC concentration to a higher degree than the mineral particulate matter.

The widest variability of the HC concentration is characteristic for the Ob Bay–Kara Sea section (the Ob river's marginal filter, Fig. 1): 6–291 μg/l for the AHC_d and 10–310 μg/l for the AHC_p (Table 1). The fluctuations in the AHC content are the greatest at the salinity within 0.05–9.3 psu (Fig. 2a), which conforms to the summer conditions of the marginal filter in the Arctic seas [7]. In Ob Bay, the surface waters contain lipids and AHC mainly in the dissolved form. Further

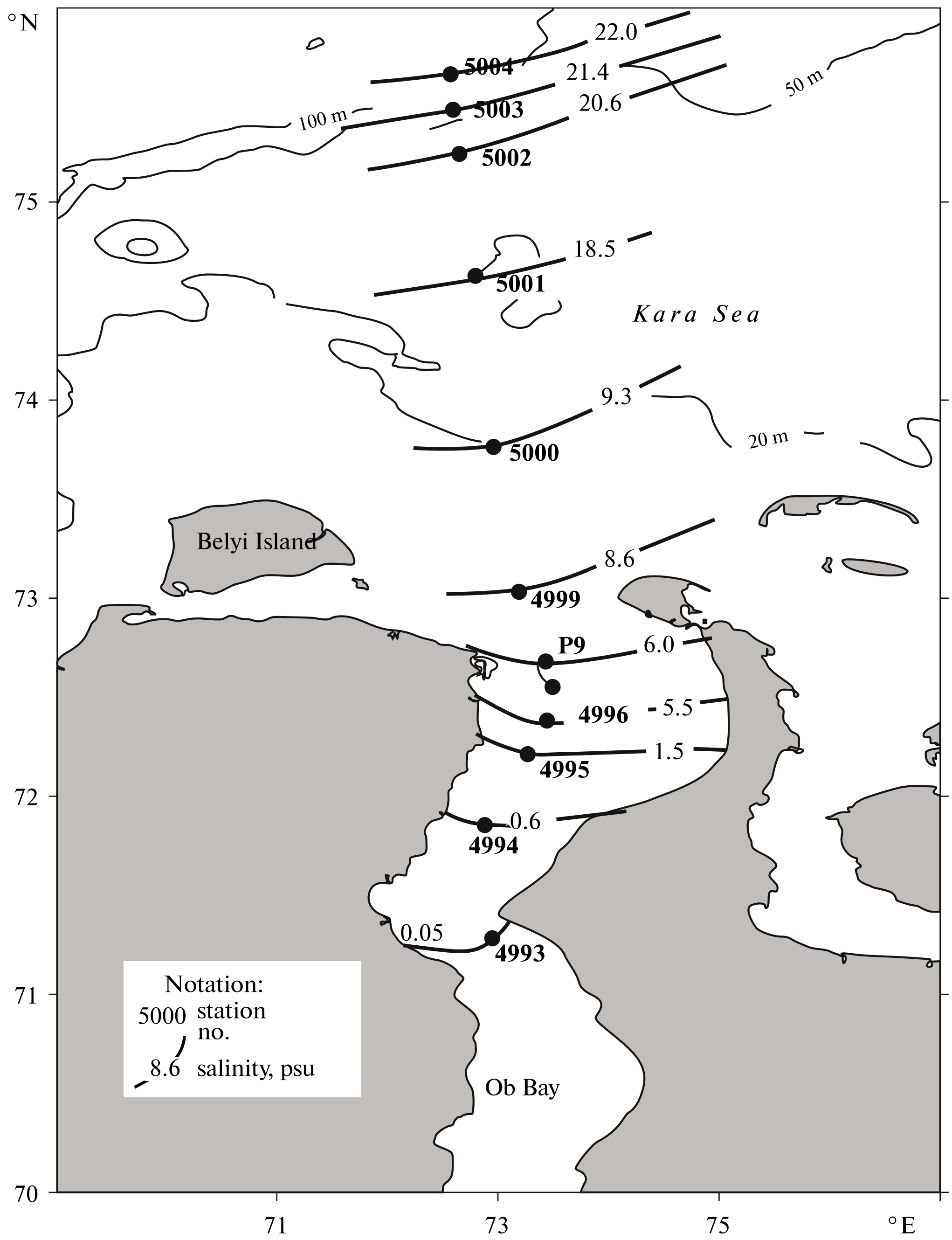

Fig. 1. The sampling scheme of the Ob Bay–Kara Sea section.

over the section, the concentrations of both the dissolved and particulate OC forms increase. At the salinity within 0.05–0.6 psu, the AHC_d/AHC_p ratio varied from 1.65 to 1.35. The water stratification is not pronounced in this area because the values of the near-bottom and surface salinity were practically the same over the section beginning at up to 72.4°N. However, the AHC_p were prevailing in the near-bottom waters (AHC_d/AHC_p values from 0.56 to 0.32), probably, owing to the effect of the nepheloid near-bottom currents [1]. The content of AHC_p in the near-bottom water taken from the Neimisto tube was here over 700 μg/l.

Within the salinity range from 1 to 5.5 psu, the bulk of the OC is removed from the surface waters; therefore, the concentrations of both the dissolved and particulate forms decrease. In this part of the section, the dissolved OC are also trapped under the particulate matter sedimentation. With the salinity growth, the concentrations of the dissolved OC decreased and those of the particulate OC increased, and, at S = 6 psu (station P9), the minimum of the dissolved forms coin-

Table 1. The content of organic compounds in the particulate matter of the surface waters of the Ob Bay–Kara Sea section

Station	S, psu	Particulate matter*, mg/l	Lipids	AHC	AHC, mg/mg of particulate matter	AHC, µg/mg POC**	AHC_d/AHC_p	PAH, ng/l	PAH, ng/mg of particulate matter
			µg/l						
4993	0.05	231.3	112.7	45.7	1.98	49.7	1.65	2.5	109.4
4994	0.6	343.7	427.0	215.9	6.28	145.0	1.35	17.2	500.4
4995	1.5	217.0	224.2	151.6	6.99	214.8	0.94	15.5	713.4
4996	5.5	196.0	279.1	189.4	9.66	291.4	0.48	3.0	154.1
P9	6	183.0	200.1	130.5	16.88	–	0.18	5.2	284.2
P10	8.6	128.0	189.4	90.8	9.59	–	0.53	5.0	387.5
5000	9.3	4.0	36.8	9.70	24.25	49.0	0.74	2.5	6175.0
5001	18.5	3.1	31.5	15.9	51.38	135.3	0.42	2.7	2387.1
5002	20.6	5.6	24.8	12.5	22.39	83.5	0.48	0.7	767.9
5003	21	5.5	35.9	20.6	37.45	170.0	1.01	0.4	763.6
5004	21	4.3	40.3	21.4	49.8	142.5	1.29	0.4	720.9

Notes: * The data on the particulate matter by Kravchishina
** The data on the POC by Belyaev.

cided with the maximum of the particulate matter (for $AHC_d/AHC_p = 0.18$), because the dissolved OC were transformed into the particulate form, and the AHC_p content per mg of particulate matter increased by a factor of 8.5 (from 2 to 16.9 µg/mg of particulate matter). This is due to the presence of large amounts of particulate matter, humic and fulvic acids, iron, and bacteria in the freshwater part of the marginal filter [7]. Moreover, the salting-out effect of the seawater causes the transformation of the dissolved OC into the particular form. In the more saline waters, the dissolved and particulate forms of these compounds varied synchronously.

In the open areas of the Kara Sea, the AHC_p content was 13 µg/l or below with concentrations as low as 6–7 µg/l near the St. Anna Trench. In the pelagic areas, the AHC_d content usually decreases to 10 µg/l (the Arctic Ocean waters, Table 2). This is evidently caused by the low nutrient content and water temperatures, which determine the low primary production in these aquatic areas [22]. Only with drawing near the coast in Blagopoluchiya Bay (the Novaya Zemlya Islands) does the AHC_d content increase to 27.8 µg/l. The existence of the circumcontinental zonality causes the increase of the AHC concentrations in the coastal shelf waters [11].

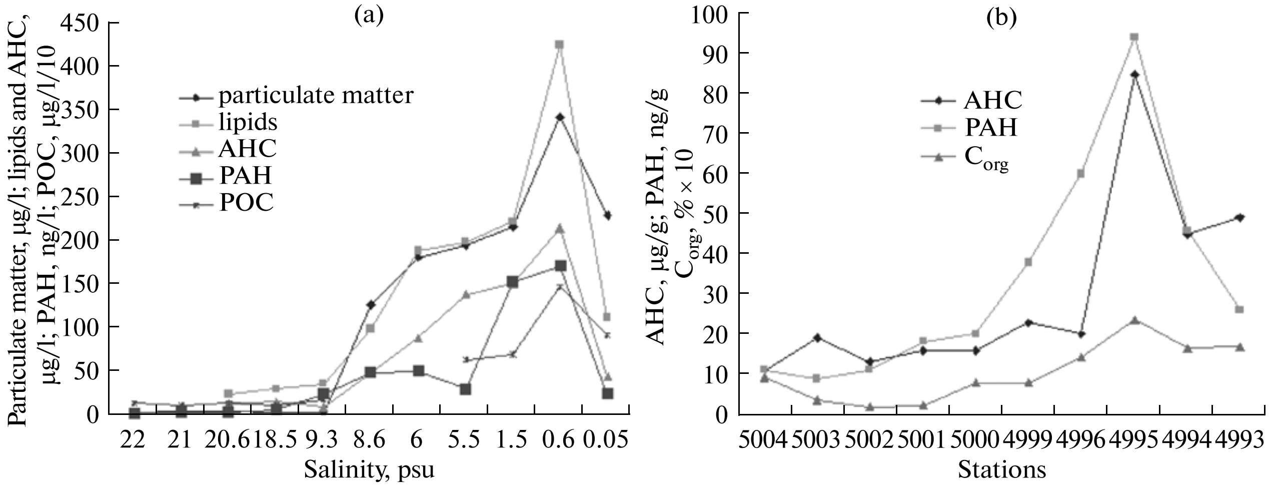

Fig. 2. The distribution of the particulate matter, the POC, the particulate forms of the lipids, the AHC, and the PAH in surface waters (a) and of the C_{org}, AHC, and PAH in the surface layer of the bottom sediments (b) in the Ob Bay–Kara Sea section.

Table 2. The content of aliphatic hydrocarbons (the IR technique) in the surface waters of the Arctic seas

Region	Year	Number of samples	Concentration, μg/l		
			average	range	standard deviation
White Sea	2001	32	12	5–45	5
	2003	13	18	11–30	6
Baltic Sea	1983–1991	285	12	1–77	21
	2008	30	11	0–49	14
Barents Sea	1998	21	15	0–22	7
	2007	6	16	8–19	4
North Sea	2001	9	22	16–36	8
West Europe shelf	2001	6	9	1–18	7
	2003	10	16	6–39	10
Northern Sea Route (the Barents, Kara, Laptev, and East Siberian seas)	2000	33	17	0–33	7
Arctic Ocean (170°E–170°W; 81–82°N)	2000	18	10	0–26	7
Kara Sea	2007	14	16	5–31	8

The PAH content decreased as well in passing from the riverine to the marine waters in the Ob Bay–Kara Sea section (Table 1). The maximum PAH concentration, as that of the AHC, was registered at 1.5 psu salinity (Fig. 2a); however, the maxima of the PAH distribution is less pronounced in the more saline waters. This is evidently caused by the different nature and sorptive abilities of these hydrocarbon classes depending on their physicochemical properties and capability for intermolecular and sorption interactions: the AHC are formed under biosynthesis and they are supplied to the basin with oil contamination [24]. The problem of the biogenic synthesis of polyarenes is still debatable, and pyrolitic processes are considered as primary PAH sources [14, 21]. Hydrophobic AHC are mainly sorbed by the particulate matter, and the more hydrophilic polyarenes with prevailing three- and four-cyclic compounds in their composition are transformed into the soluble form. However, as for the AHC, the concentration of the particulate matter remains a controlling factor in the PAH distribution: r (particulate matter–PAH) = 0.77 (n = 9). There is a correlation between the distribution of these hydrocarbon classes. At the same time, the maximum content per mg of particulate matter was found at the seaward stations; i.e., the concentrations of the polyarenes decrease to a lesser degree compared to the particulate matter.

In the PAH composition, the phenanthrene produced by natural processes was prevailing. In Ob Bay, the fraction of perylene is significant and is also of biogenic origin [14, 28]. The minor components of the PAH composition in the water and filtrational particulate matter include benz(a)pyrene, which is characterized by carcinogenic and mutagenic properties.

In the mouth areas of the Ob and Yenisei rivers, complicated relationships between the distribution of the salinity, particulate matter, and OC were also previously registered (in 1993 and 1997 [5, 21]). The influence of phenomena such as the tidal activity, the near-bottom layer roiling, and the storm effects disturbs the correlations between the distribution of the particulate matter and the OC. Moreover, new OM formation owing to the carbon bioassimilation is also possible, which was found in the Yenisei River mouth according to the ^{13}C isotope composition [3, 5]. At the seaward stations of the marginal filter, the OM content increased due to two main sources: the OM of the marine plankton and the colloidal and the dissolved and coagulated OM transformed partially into particulate matter of >0.4–0.6 μm grain size in the mixing zones [6].

In the bottom sediments, in their surface layer, the concentration of C_{org} varied from 0.01 to 2.04% (0.90% on average, $\sigma = 0.52$) with the AHC content being from 3.3 to 76.6 μg/g of the dry mass (35.3 on average, $\sigma = 25.6$, $n = 32$) and 0.02 to 3.35% of the C_{org} (1.67% on average) (Table 3).

In the Kara Sea–Yamal Peninsula section, seawards from the coasts, a typical view of the distribution of the bottom sediments on a slope was traced: the coarse-grained fractions were superseded by those of finer grain size. The change of the sediment types followed the following scheme: sands—coarse siltstones—fine-silty oozes—silty-clayey oozes (Table 3). The capability for the sorption of HC, as well as of OM, in general, depends on the dispersion of the sediments [11, 16]. Therefore, the correlations were found between the moisture M (depending on the lithological type of the sediment), the OM, and the

AHC: $r(C_{org}–M) = 0.86$, $r(C_{org}–AHC) = 0.94$, and $r(M–AHC) = 0.74$ ($n = 9$). Because of their high hydrophobicity and their rather low solubility, HC are easily sorbed on particulate matter and, under its sedimentation, get into the bottom sediments. In case of the local supply of anthropogenic HC or newly formed OM, these relations are usually disturbed [11].

Within the C_{org} composition, the maximum AHC fraction is characteristic of the sandy sediment of station 4955 located on the slope near to the Yamal Peninsula. This might be caused by the passive HC sorption by the coarse-grained sediments at shallow depths [11]. Because of this, despite the lower concentrations per air-dry mass, the AHC fraction in the sandy sediments is usually higher than that in the oozes. It is remarkable that, in the samples collected with a Neimisto tube and an Okean dredge, the values of the compounds considered were practically unchanged (station 4957, Table 3).

In the Ob Bay–Kara Sea section (Table 3), the shallow-water sediments of the corner part consisted of black and fine-silty oozes (stations 4993 and 4995–5000, respectively). Coarse-grained siltstones and medium-grained sands were found at the periphery stations (nos. 5001 and 5002, respectively). Silty–clayey oozes were found only at the deepwater station 5004. The variability of the OC content in the surface layer of the sediments over the section is rather wide: from 0.20 to 2.36% for the C_{org}, from 3.7 to 42.4 μg/g for the AHC (0.04–0.21% of C_{org}), and from 11 to 94 ng/g for the PAH (0.12 to 0.84×10^{-3}% of C_{org}). The OC maxima are related to station 4995 (Fig. 2b) located in the physicochemical area of the marginal filter at the zone of avalanche sedimentation. Here, with the change of the colloidal particles' charge (S = 1.5 psu), the particulate forms of the OC (their concentration maximum, Fig. 2a) pass to the underlying waters and sediment. These data are close to those obtained formerly when the OC content in the bottom sediments decreased irregularly over the Ob Bay–Kara Sea section [13, 19, 21]. In the corner part of Ob Bay, the HC fraction in the OM's composition is decreased compared to the seaward part of the section and amounted only to from 0.12 to 0.4×10^{-3}% for the AHC and PAH, respectively, at station 4995. Because of this, the HC may be considered as a minor component of the precipitating OM. Most probably, this is caused by the fact that the HC fraction in the aquatic areas distant from industrial centers and in the newly formed OM is lower than that in the transformed OM and in the impact areas [11]. Therefore, while the AHC and PAH content at station 4995 amounted to 1.8 μg/mg of C_{org} and 1.5 ng/mg of C_{org}, respectively, the concentrations at station 5002 increased to 2.7 μg/mg and 5.5 ng/mg, respectively. Beyond the bounds of the marginal filter, the HC fraction in the pelagic sediments decreased anew to 0.05 and 0.12×10^{-3}% for the AHC and PAH, respectively.

Correlations of the high coefficient values are found for the C_{org} and AHC concentrations: $r(AHC–PAH) = 0.75$, $r(AHC–C_{org}) = 0.87$, and $r(PAH–C_{org}) = 0.81$ ($n = 10$). The correlations between the lithological type of sediments and the OC are weaker: $r(M–C_{org}) = 0.76$, $r(M–AHC) = 0.51$, and $r(M–PAH) = 0.57$. Because of this, the OC concentration is determined not only by the material component of the sediments but also by the processes in the Ob River's marginal filter. The degree of the concentration increase at station 5003 might be caused by the variations in the sediment grain-size type or by the HC sorption in the biological part of the marginal filter.

In the PAH composition of the surface layer of the sediments in the Kara Sea, phenanthrene is prevailing with the fraction being from 65% in Ob Bay to 100% in the Kara Sea sediments (Fig. 3). It is thought that he phenanthrene is of natural origin [14, 21, 30]. This arene is formed in relatively unpolluted regions at OM transformation, either under diagenetic processes in humus-rich sediments (an indicator of humus accumulation process) or under dehydrogenation of steroids by microorganisms [21, 30]. In the corner part of Ob Bay (station 4995), the prevailing compounds include naphthalene (25% of the total PAH). Here, the naphthalene–phenanthrene ratio is over 1, which may point to the presence of pyrogenic compounds [19, 26]. In the sediments of station 4995, the fraction of fluoranthene and pyrene is also increased (64% of total PAH), which, with quite a low fluoranthene/pyrene ratio (0.6), may point to the presence of pyrogenic compounds. The prevalence of anthropogenic polyarenes over natural compounds is also revealed in the quite low (for an estuarine zone) perylene fraction (<20%), which usually indicates terrigenous OM [14, 28]. The maximum perylene fraction (50%) is associated with the sediments of station 4996 with the maximum Eh values over the entire section (+476) [15]. A change of the redox conditions of the sedimentation affects the formation of this polyarene in the sediments. The most probable perylene sources are peat decomposition products transferred by the Ob River [21].

The differentiation of the sediment substance in the zones of increased hydrodynamic activity causes the precipitation of coarse-grained matter at the depths shallower than 100 m and of ooze sediments in the deep trenches and depressions with bottom lowering. The burial depth and the variations in the material's composition and the redox conditions of the sediment determine the irregular changes in the HC concentrations, which are especially pronounced at the transition from the upper oxidized to the subsurface reduced layer. The content of AHC in these layers usually decreases even faster than that of the OM [11]. This effect is ascribed to the changes in the rates of the hydrolysis and redox processes in the sediment mass [18].

The most pronounced changes in the AHC content within the bottom sediments' mass were registered at station 4970 located at the Novaya Zemlya Trench in

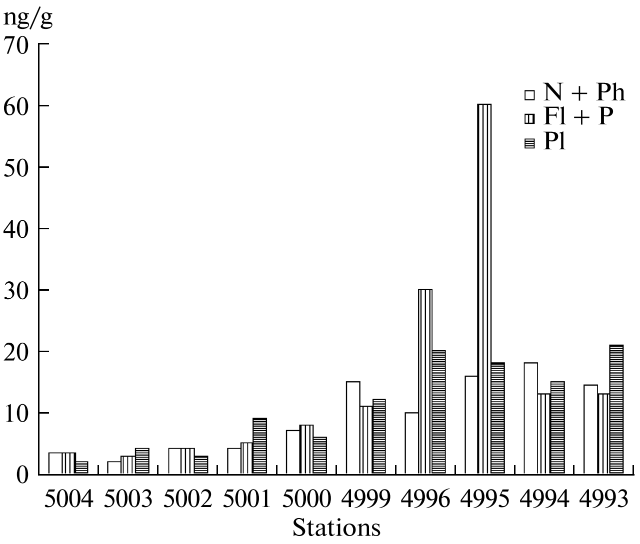

Fig. 3. The distribution of the PAH (ng/g) in the surface layer of the bottom sediments in the Ob Bay–Kara Sea section.

the zone of intense sedimentation. Here, the surface layer (0.5–14 cm) was sampled with a Neimisto tube, and an LDT was used to sample the layer of 10–58 cm. The oxidized layer of sediments amounted to about 30 cm; the Eh values varied from +604 to +120 in the 0–5 and 50–58 cm layers, respectively [15]. In the sediment's mass, a few maxima were seen in the OM and AHC distribution, which coincided with the variations of the redox conditions and the material composition of the sediment (the layers of 10–20 and 39–41 cm, see Fig. 4). The strongly oxidized medium caused the decrease of the AHC content by a factor of almost 12 from the 0.5–1 to 7.5–12.5 cm layer; the content of Mn decreases by a factor of 17.7 [15]. In the surface layers of the sediments, under contact with the oxygen of the near-bottom water, the processes of aerobic oxidation are the most intense, which is shown by the great abundance of oxic microflora cells occurring in the sediments [2]. Later, with the burial depth, below the boundary of the oxygen presence, the OM is transformed far slower, and even a small increase of the AHC concentrations is seen. The maximum of the AHC concentrations at the layer of 39–41 cm accompanied by a C_{org} decrease is probably caused by the anoxic decomposition of the OM. Because of this, a correlation with the burial depth distribution of these compounds is absent: $r(\text{AHC}–C_{org}) = –0.087$.

At station 4974 located at the slope of the Novaya Zemlya Trench, the concentrations of the AHC and C_{org} were lower and varied to a lesser degree: from 3.3 to 8.0 μg/g and from 0.99 to 1.26% of C_{org} (Fig. 4). Here, a few maxima were also seen in the AHC distribution. The first maximum is situated in the 1–2 cm layer, where the Eh values decreased from +497 to +150; the second one (more pronounced) was at the layer of 30–35 cm, where Eh variation also took place (from +30 to +74). A similar distribution of the AHC and C_{org} was observed at the slope of the Franz–Victoria Trench in the northern part of the Barents Sea [11]. The analysis of the alkanes in the sediment mass of the Franz–Victoria Trench showed an increased content of n-C_{20} and n-C_{22} homologues; i.e., the HC formation proceeded owing to the microbial destruction of the OM. By analogy with the White Sea, one may suppose that oxic processes proceed intensively only in oxidized sediments. Particularly, in the sediment mass of the White Sea Basin, the intensity of the sulfate reduction in the boundary layers differed by a factor of 70 (901 and 13 μg of S/dm^3 day) [18]. Under the reduced conditions in the sediment mass, the microbial dehydrogenation of various steroids may also cause an increase of the HC content [27]. In the deep-seated layers of the more transformed sediments, where the abundance of bacteria is usually sharply decreased [5, 18], both the AHC and C_{org} concentrations decreased; therefore, the AHC fraction in the C_{org} composition was practically invariable.

In the sediment mass of the Ob Bay–Kara Sea section, the AHC and C_{org} concentrations decreased synchronously (the correlation coefficient varied within 0.54–0.94). At station 4993, the black ooze was only slightly compacted with depth (0–35 cm layer); the parameters considered varied from 0.71 to 64.5 (moisture), from 25 to 17 μg/g (AHC), and from 1.68 to 1.72% (C_{org}). The Eh values in the sediment mass remained constant (up to +20 mV) [15]. However, even at station 4995 in the area of avalanche sedimentation, the AHC content in the 0–14 cm layer almost threefold decreased from 42.4 to 14.9 μg/g with the minimum of 11 μg/g in the layer of 2–4 cm, where the transition from oxidized to reduced sediments was registered. Evidently, under rapid burial of fresh OM, intermediate zones of suppressed microbiological utilization and resyntheses are formed. This causes the appearance of microfacies of fresh HC composition with no visible changes of the initially precipitated organic matter [31].

In conclusion, one must note that the rather low HC values in the surface seawaters of the open areas of the Kara Sea (on average, 15.5 μg/l of AHC_d and 14.1 μg/l of AHC_p) point to their minor supply from anthropogenic sources. Their content falls into the range of concentrations for the Arctic seas (10–22 μg/l, Table 2). These values, as those obtained formerly [3], are below the fishery values of the maximum allowable concentrations for oil HC in aquatic basins (50 μg/l). Towards the pelagic area, a pronounced decrease of their concentrations is registered. The good correlation between the concentration of lipids, HC, C_{org}, and particulate matter allows one to suppose that, as formerly [22], the erosion of the coasts and riverine runoff are the main sources of the AHC.

At the river–sea boundary, a drastic transformation of the HC supplied with the rivers takes place [11]. In

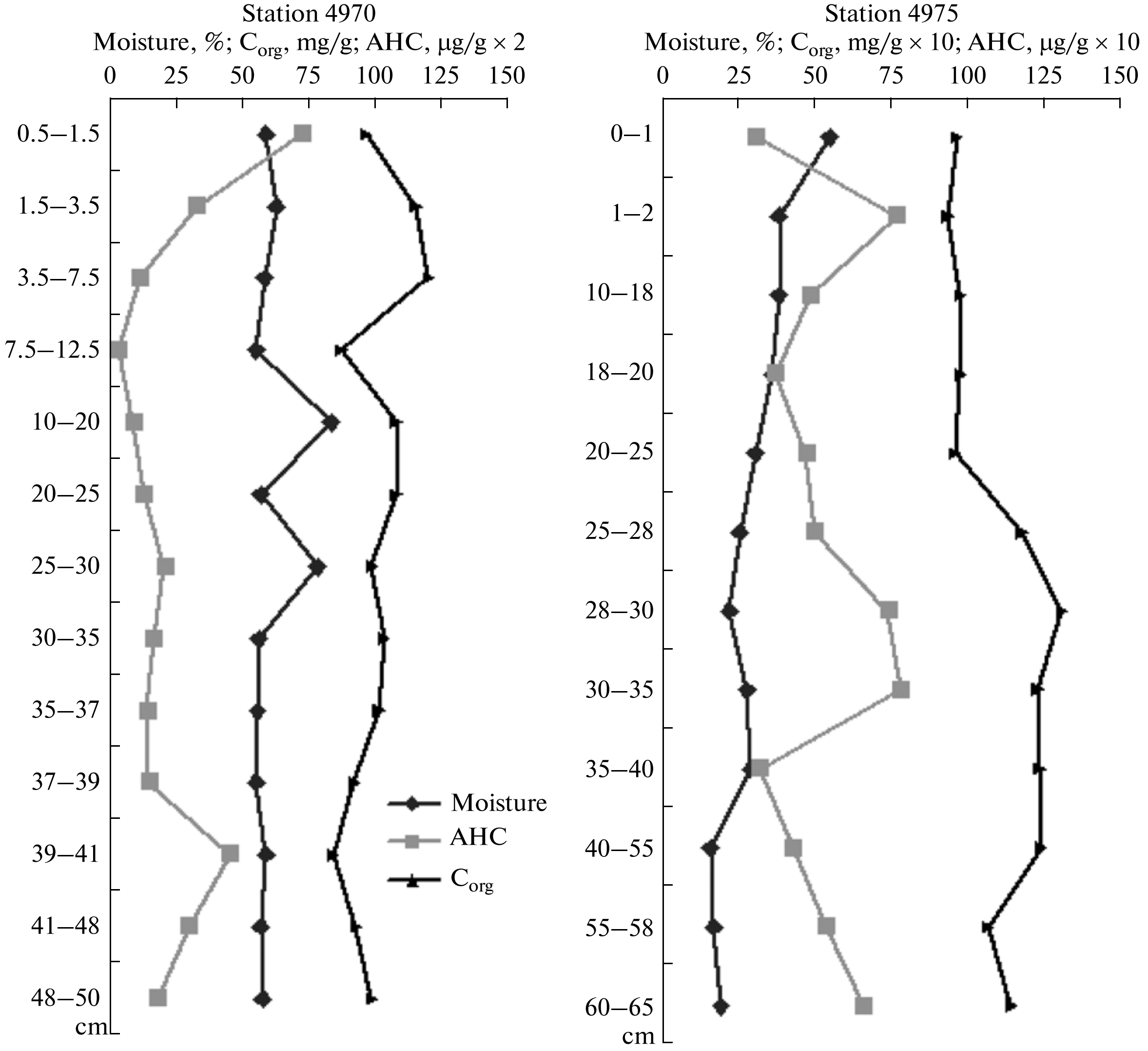

Fig. 4. The distribution of the moisture, AHC, and C_{org} in the bottom sediment mass.

this case, two regularities are of fundamental importance: the bulk of the sediment matter is precipitated near the river mouths, and this sedimentation is determined by the salinity in the zone of the riverine and marine water mixing [6]. Because of this, the variability of the OC concentrations at the Ob Bay–Kara Sea section is controlled by the variations of the salinity and particulate matter.

According to the hydrological data, the sharpest front is related to 72.20°N, i.e., to the region of 1.5 psu salinity (near station 4995, Fig. 2a). The front's location is represented in the spatial distribution of not only the HC but also of the particulate matter, fluorescence, and chlorophyll *a* [1]. In the more saline waters, the amount of OC in the particulate matter increased anew owing to flocculation. At $S > 9.3$ psu, the content of OC_d and OC_p decreased. In the surface waters, the concentrations of the particulate matter, POH, lipids, and AHC_p decreased synchronously: *r*(lipids–particulate matter) = 0.78 and *r*(AHC–particulate matter) = 0.74 (Fig. 2a). In the area of the riverine and marine water mixing, the content of AHC_p is higher than that of AHC_d. Only at the marine stations does the AHC_d anew prevail. Over the section in general, the difference of the concentrations between the riverine and marine waters for the AHC_d was higher than that for the AHC_p. Such a distribution between the dissolved and particulate AHC forms is characteristic for the Arctic rivers flowing over humus-rich marshy soils [6]. Because of this, not only in the Ob River mouth but also in other riverine mouths (particularly, in that of the Severnaya Dvina River [12, 23]), the concentration losses are lesser for the AHC_d than those for the AHC_p (Table 4). On the contrary, the rivers of the temperate and tropical zones are characterized by a higher degree of their AHC_p content variations at the river–sea geochemical barrier.

Table 3. The content of organic compounds in the surface layers of the bottom sediments

Station	Latitude, N	Longitude, E	Depth, m	Sampling tool	Layer, cm	Dediment description*	Moisture, %	C_{org}, %	AHC, μg/g	PAH, ng/g	% of C_{org}	
											AHC	PAH × 10^{-3}
4946	71°53.8	60°56.4	136	Dredge	0–3	SCS	60.8	1.26	75.9	51	0.42	0.41
4948	71°45.40	62°04.30	134	Dredge	0–2	FSS	54.5	1.11	76.6	44	0.47	0.40
4950	71°39.28	63°09.18	119	Dredge	0–2	FSS	42.1	0.90	64.1	76	0.49	0.84
4952	71°27.5	64°14.73	96	Dredge	0–2	FSS	49.3	0.73	66.4	10	0.62	0.14
4955	71°12.22	66°14.58	17	Dredge	0–2	CS	19.5	0.01	4.6	32	2.88	29.09
4956	71°14.95	65°51.73	30	Dredge	0–2	FSS	23.4	0.08	28.0	54	2.57	7.20
4957	71°17.62	66°46.59	38	Dredge	0–2	FSS	43.8	0.52	33.2	19	0.44	0.36
4957			38	Neimisto tube	0–2	FSS	56.2	0.51	31.0	15	0.42	0.29
4958	71°20.81	65°24.25	111	Dredge	0–2	FSS	64.3	1.29	37.7	34	0.20	0.26
4960	71°25.06	64°55.20	119	Neimisto tube	0–1	FSS	67.0	1.07	40.3	31	0.26	0.29
4969	75°38.72	63°38.67	39	Dredge	0–2	FSS	33.9	0.53	11.3	40	0.15	0.76
4970	75°21.85	64°13.73	360	Neimisto tube	0.5–1.5	SCS	59.0	0.97	36.1	14	0.26	0.14
4971	75°38.70	63°43.19	178	Dredge	0–2	FSS	35.8	0.53	3.3	33	0.04	0.63
4972	75°37.97	63°42.95	85	Dredge	0–2	FSS	38.8	0.94	3.9	26	0.03	0.28
4974	75°34.82	64°09.31	170	Dredge	0–1	SCS	57.0	0.99	3.3	12	0.02	0.12
4975	75°38.76	64°03.94	100	Dredge	0–1	GS	40.7	0.68	7.0	33	0.07	0.49
4979	75°57.78	66°18.72	40	Dredge	0–3	SCS	67.9	0.97	4.9	87	0.03	0.90
4983	76°55.49	70°19.92	510	Neimisto tube	0–1.5	FSS	69	2.04	6.3	10	0.02	0.05
4988	70°42.72	70°57.80	213	Dredge	0–2	FSS	57.5	0.94	8	64	0.06	0.68
4990	76°8.03	72°35.71	106	Dredge	0–2	FSS	54.7	0.80	4.5	17	0.04	0.21
5003	75°26.5	72°29.8		Dredge	0–1	SCS	30.5	0.36	9.5	9	0.18	0.25
5004	75°34.33	72°24.80		Dredge	0–2	SCS	56.3	0.92	5.4	11	0.04	0.12
4993	71°14.90	72°51.40	20	Neimisto tube	0–0.5	BS	70.1	1.68	25.3	26	0.01	0.01
4994	71°44.62	72°47.38	16	Neimisto tube	0–0.5	CS	72.9	1.67	22.6	46	0.09	0.28
4995	71°44.01	72°47.38	16	Neimisto tube	0–1	FSS	82.2	2.36	42.4	94	0.12	0.40
4996	73°49.16	72°47.38	15	Neimisto tube	0–1	FSS	70.5	1.44	10	60	0.05	0.42
4999	72°57.63	73°17.89	27	Dredge	0–1	FSS and C	45.5	0.79	11.4	38	0.10	0.48
				Neimisto tube	0–2	FSS and C	65	0.84	3.7	20	0.03	0.24
5000	73°45.19	72°35.77	28	Dredge	0–2	FSS and C	85.6	0.63	8	21	0.09	0.33
5001	74°35.56	72°46.14	23	Dredge	0–1	CS	27.3	0.21	6.5	18	0.21	0.84
5002	75°08.8	72°35.7	36	Dredge	0–2	C	21.5	0.20	5.4	11	0.19	0.55

* SCS is silty–clayey ooze, FSS is fine-silty ooze, CS is coarse-grained siltstone, GS is gray ooze, BS is black ooze, FSS and C are fine-silty oozes with an increased content of clayey matter, and C is claystone.

Table 4. The losses of AHC at the river–sea geochemical barrier

River	Layer	Losses*	River	Layer	Losses
Ob [21]	surface, 1993	–/25	Neva	PMC	47/20
Ob	surface, 2007	37/27		surface	63/85
Severnaya Dvina			Neman	surface	75/–
	surface, 2001	25/0	Amur	surface	10/75
	surface, 2003	33/14		bottom	50/–
	surface, 2006	24/15	Daugava	PMC	68/48
	surface, 2007	–/67		surface	88/80
Yenisei [21]	surface, 1993	–/17		bottom	77/80
Congo	surface	47/73	Narva	surface	17/80
Limpopo	surface	68/92	Dniepr	surface	84/80
Zambezi	surface	65/82	Rhone [20]	surface, 1992	–/85
				surface, 1994	–/76

* The numerator is the dissolved and the denominator is the particulate matter.

The studies of the PAH behavior in the estuarine zones showed that the submicron fractions contained mainly light polyarenes [20]. At the outer boundary of the shelf, the colloidal fraction may be somewhat enriched in compounds of higher masses, because of their redistribution from the aquatic phase under flocculation. The low PAH concentrations in the particulate matter may be caused by the high flow rate of the riverine waters and by their high dilution [21, 25]. In the substance from the sedimentation traps, the relative enrichment of the particulate matter (especially of the fecal pellets) compared to the underlying sediments was found for phenanthrene, fluoranthene, and pyrene [25]. The selective transition of the light polyarenes from the dissolved form into the particulate matter, either by sorption and coprecipitation or by bioaccumulation and biosedimentation, is assumed.

The surface layer of the bottom sediments considered contained 2.36 or below of C_{org}, 76.6 μg/g or below of AHC, and 94 ng/g or below of PAH; i.e., the Kara Sea's bottom sediments were characterized by rather low concentrations of OM and HC. Like values were found for the Kara Sea sediments in 1993, when the average C_{org} concentration amounted to 1.5%, while increasing to 1.9% in the mixing zone and decreasing to 0.7% in the open parts of the Kara Sea [22].

The most intense sedimentation of suspended particles is seen in the Ob River's marginal filter; hence, the maximum concentrations of the particulate matter are associated with this area. The character of the HC composition depending on the sources (allochthonous, autochthonous, or anthropogenic), the redistribution between the migratory forms in the biogeochemical processes, and the sedimentation conditions (the area of avalanche sedimentation) disturb these regularities. It was also formerly noted that no pronounced decrease of the main OM components in the bottom sediments proceeded towards the central part of the sea, which is caused by the OM's nature formed by the terrigenous plant matter [22]. Therefore, whereas there is a correlation between the sediment's moisture and the C_{org} for all the treated areas of the Kara Sea ($n = 30$), the relationships between the AHC concentrations, moisture, and C_{org} are quite weaker: $r(\text{M}–C_{org}) = 0.86$, $r(\text{M–AHC}) = 0.37$, and $r(C_{org}–\text{AHC}) = 0.36$. Evidently, the AHC content is affected not only by the lithological type of the sediment but also by the conditions of the sedimentation.

In the ooze and sandy bottom sediments, the AHC concentrations of 100 and 10 μg/g, respectively, are considered as the minimum values under which no biological effects are present or appear as reversible reactions of marine organisms [3, 11]. In 2002–2003, during the drilling works in Ob Bay, the AHC concentration in the surface layer of the sediments increased to 55–205 μg/g [3]. The AHC decrease to the time of our studies (the maximum concentration of 42.4 μg/g) might point to the decomposition of oil HC or to the burial of bottom sediments with newly supplied and relatively pure matter. The rate of the sedimentation for the Ob Bay aquatic area amounts to 0.1–0.13 cm/year [3]. The sedimentation of the particulate matter at the Ob River's mouth is from 200 B at the salinity of 1–3 psu to 1700–26000 B at 5.5–9 psu salinity at the depot's center [7] (B is Bubnov's unit of the sedimentation rate, mm/1000 years). The flux of C_{org} also reaches its maximum values of about 27 mg of C/m^2 day. Hence, the distant transfer of pollutants supplied with the surface and underground wastewaters from drilling and technological localities with the water currents crossing the oil recovery areas has become a more important process in the bottom sediment's contamination. As a result, the AHC content in Taz Bay is higher than that in Ob Bay owing the waters flowing over the Pur oil fields [3].

Table 5. Concentrations of PAH (ng/g) in the surface layers of the Arctic bottom sediments

Region	PAH number/method	Year	Concentration	Reference
Barents Sea				
Northwest	11/GC–M	1992	200	[32]
Southeast	11/GC–M	1992	230	[32]
Coasts of the Novaya Zemlya Islands	11/GC–M	1992	97*	[32]
White Sea				
The whole sea	27/GC–M	1994	13–208	[26]
Basin	8/HPLC	2001	1–44	[23]
The Severnaya Dvina River's mouth	8/HPLC	2003	11–140	[12]
The Severnaya Dvina River's mouth	8/HPLC	2005	120–227	[13]
Dvina Bay	8/HPLC	2006	12–89	[13]
Kara Sea				
Northwest	12/GC–M	1993	46	[21]
Ob River delta	12/GC–M	1993	54	[21]
Yenisei River delta	12/GC–M	1993	84	[21]
Sea shelf	27/GC–M	1993	166	[19]
Ob River delta	27/GC–M	1993	232	[19]
Yenisei River delta	27/GC–M	1994	537	[19]
Baidaratskaya Bay	27/GC–M	1993	77	[19]
Northwest	27/GC–M	1993	102	[19]
Ob River delta	8/HPLC	1991, 1995	15–47	[14]
Yenisei River delta	8/HPLC	1995	40–20	[14]
Yamal section	10/HPLC	2007	37	this study
Northwest	10/HPLC	2007	30	
Ob River delta	10/HPLC	2007	40	
Laptev Sea				
Lena River delta	11/GC–M	1993	69	[21]
Southwest	11/GC–M	1993	97	[21]
Pacific Ocean (Alaska Inlet)				
Cook Inlet	20/GC–M	1976, 1979	10–300	[29]
Central part	20/GC–M	1976, 1979	100–200	[29]
Cape Kodiak	20/GC–M	1976	6–100	[29]
Bering Sea				
Southwest	20/GC–M	1975	3–20	[29]
Norton Sound	20/GC–M	1976, 1977	1–20	[29]
Navarin Bay	20/GC–M	1980	30–40	[29]
Beaufort Sea				
Alaska shelf	20/GC–M	1976	200–300	[29]
Mackenzie River delta	11/GC–M	1986, 1987	980	[32]
Mackenzie River shelf	20/GC–M	1986, 1987	860	[32]
Shelf edge	20/GC–M	1986, 1987	500	[32]
Pelagic area	20/GC–M	1986, 1987	550	[32]

* With a single value, the average concentration is given.

The concentration of the PAH is determined to a considerable degree by the quantity of identified individual polyarenes. Therefore, their concentrations in the bottom sediments varied in a wide range for the different Arctic regions (Table 5). However, the ratios of the primary markers remained quite steady. Particularly, for the Ob River mouth, the Fl/(Fl + P) ratio, according to the author's and published data [19, 21], was below 1, which might point to a minor supply of pyrogenic polyarenes. In general, just as previously, the PAH are the products of the transformation of detritus matter and solid runoff, because phenanthrene is prevailing in their composition (up to 78% of the total PAH). Pyrolytic sources are negligible in the Arctic. To the end of the summer, the flux of anthropogenic PAH supplied via the atmosphere from the Eurasian region, Eastern Europe, and Russia is decreased [21]. In the winter, aerosols from the low-latitudinal areas are precipitated onto the ice and then transferred by the rivers to the seas.

The anthropogenic HC are decomposed quite rapidly, even at low temperatures [11, 14]. In particular, the microbial destruction of benz(a)pyrene in the surface seawaters amounts to ~400 t/year, i.e., ~8% of the total BP supply from the natural and anthropogenic sources [14]; thus, the natural HC are prevailing in the water, particulate matter, and bottom sediments. However, in the corner part of Ob Bay (station 4993), the PAH composition shows the features characteristic for oil and pyrogenic compounds. Hence, the effect of the anthropogenic PAH is revealed in the abiotic area of the marginal filter; beyond it, the PAH composition is stabilized.

In the Arctic seas, despite the low temperatures, the AHC decomposition and formation registered in the bottom sediment mass is comparable to other climatic zones [11, 23]. Nearby trenches, because of the high hydrodynamic activity of the waters and the erosion of the underlying masses, the sedimentation may include the OM of various genesis [16], which causes variations of the AHC and C_{org} concentrations within the sediment mass. In this case, the main role in the ways of the transformation of the HC and the total OM is played by the microbial oxidation and sulfate reduction [5].

CONCLUSIONS

The content of HC in the surface waters, on the average for the AHC_d, was 16 μg/l for the entire sea and 8 μg/l in the open waters, which conforms to their level in the surface seawaters. These parameters are considerably below the fishery values of the maximum allowable concentrations for oil products in aquatic basins, which may testify either to a minor supply of oil pollutants or to their rapid degradation. The PAH composition with prevailing phenanthrene also points to the natural HC genesis.

The surface layer of the bottom sediments is characterized by rather low HC concentrations: 3.3–76.6 μg/g for the AHC and 9–87 ng/g for the PAH. This is due to the relatively low OM content in the sediments (the C_{org} concentrations were 2.36% or below), which may be caused by the short vegetation time of the diatom algae and, hence, by the low annual average values of the biomass. The HC content is affected not only by the lithological type of the sediment but also by the conditions of the sedimentation.

The widest variability of the HC concentrations in the water and bottom sediments was registered at the Ob Bay–Kara Sea section. Their distribution follows the regularities of a marginal filter of Arctic rivers. Considering that no HC concentration increase in the water and bottom sediments took place in the recent years, one may conclude that the Ob River's marginal filter has coped until now with the contamination supplied.

In the course of the reductive diagenesis in the recent sediments of the Kara Sea, both the degradation and formation of HC proceed, which may point to the high rate of the HC transformation in the sediments of the Arctic seas.

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