

Features of Distribution of Hydrocarbons in Bottom Sediments of the Streletskaia Bay (Black Sea)

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Abstract

Purpose. The purpose of the study is to identify the features of spatial and vertical distribution of hydrocarbons in the bottom sediments and to assess the likely sources of their ingoing to the port coastal water area (at the example of the Streletskaia Bay).

Methods and Results. The samples of the bottom sediments surface layer (5 sampling stations) and the columns of marine sediments (2 columns) were taken during the joint expedition of the Department of Marine Sanitary Hydrobiology (FRC IBSS) and the Department of Marine Biogeochemistry (FRC MHI) in July 2021 as a part of a long-term monitoring of the Sevastopol bays. The features of spatial distribution in the sediment surface layer (0–5 cm), the profiles of vertical distribution of the geochemical characteristics of bottom sediments, hydrocarbons and n-alkanes, and the individual diagnostic indices (markers) were analyzed. The history of hydrocarbon accumulation resulted from the 50-year long human activity was considered, and the anthropogenic load on the bay water area was assessed.

Conclusions. The bottom sediments composition of the Streletskaia Bay promotes the accumulation of hydrocarbons: the aleurite-pelitic silts of high natural humidity are ubiquitous, and the organic carbon average contents in the surface layer (5.1%) and in the bottom sediments thickness (5.3%) significantly exceed the values typical of the other water areas in the Sevastopol region (1.2–3.7%). The hydrocarbon concentrations in bottom sediments ranged from 328 to 2175 mg/kg (the average value is 1160 mg/kg), that exceeds the pollution levels in many port areas of the Black Sea. The concentrations of the studied substances increase from the top of the bay to its apex. The composition of n-alkanes and the nature of chromatograms indicate a mixed origin of hydrocarbons at the dominating allochthonous (incoming from land) compounds, and also the presence of n-alkanes of the autochthonous and petroleum origin. Based on the data resulted from the chronology of hydrocarbon accumulation, the maximum anthropogenic loads on the bay fell on the periods 1967–1973 and 1985–1991. These were the years of intensive economic development of the city and the population growth in the sub-district adjacent to the studied water area in the Streletskaia Bay. In recent years, the intensity of hydrocarbon accumulation has been decreasing, but the signs of oil pollution are still present.

Keywords: bottom sediments, hydrocarbons, n-alkanes, markers, Streletskaia Bay, Black Sea

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Introduction

It is known that the marine ecosystems of the Sevastopol region bays are subject to chronic pollution [1–4]. According to the research results presented in [1], one of the typical examples of a water area influenced by long-term anthropogenic load is the Streletskaia Bay, which, in terms of pollution level of bottom sediments (BS) with petroleum hydrocarbons in the region, is in second place only after the Sevastopol and Yuzhnaya bays [2–4]. If the reduction in the naval base functioning led to a significant decrease in the amount of petroleum hydrocarbons (PHC) in bottom sediments [1], then the increase in the rate of urban development accompanied by the flow of untreated sewage and storm water entails the flow of organic substances, including those of oil origin, into the water and bottom sediments of the bay [1].

Bottom sediments sorb hydrocarbons (HC), which affects the life activity of bottom inhabitants and indicates the water area state. In addition, bottom sediments can also serve as a source of secondary water pollution, so monitoring their condition is very important. Contamination with hydrocarbons depends on the BS sorption capacity, which is determined by their chemical, mineralogical, and mechanical composition. Since BS are capable of accumulating hydrocarbons, it is important to study the concentrations of the substances under consideration on a time scale in order to understand the intensity of pollution, its dynamics, and also to find relations between pollution and anthropogenic activity in the port water area.

Therefore, pollution of the aquatic environment and BS with oil and petroleum products is one of the most important environmental problems for the Sevastopol port waters and has been systematically studied by the Department of Marine Sanitary Hydrobiology of the A. O. Kovalevsky Institute of Biology of the Southern Seas since 1973 [1–7]. In the Streletskaia Bay water area, the assessments of BS pollution dynamics over the long-term period covering 1985–2009 [7] and period of 2003–2015 concerning the concentrations of chloroform-extractable substances (CHES) and PHC [1] were carried out before. A reliable decrease in PHC concentrations in bottom sediments was revealed in 2003–2015; this trend was absent for CHES. The average concentration of CHES in the bay in 2015 was 9750 mg/kg, PHC – 3600 mg/kg. It was determined that the studied groups of substances were distributed unevenly throughout the bay: an increased content of both CHES and PHC was noted at the top and central part of the water area, and moderate content – at the exit from it [1, 7]. By contrast, in 2017, HC concentrations in the BS of the Black Sea open part southwards of the Crimean Peninsula reached 64 mg/kg [8], and in the sediments of coastal areas they varied within the range of 93–590 mg/kg off the Crimean coast and 53–270 mg/kg – off the coast of the Caucasus [9].

In the aquatic environment, a significant part of hydrocarbons consists of aliphatic hydrocarbons (AHC), in particular n-alkanes, the composition of which (despite their share of < 10% of AHC) determines the origin of hydrocarbons.

Scientific literature pays considerable attention to them [10–12]. They come from both petrogenic and biogenic sources [13, 14]. Petrogenic hydrocarbons are associated with pollution; they enter water areas during oil and petroleum product spills, as well as through seepage from bottom sediments¹. At the present stage, biogenic AHC are present in water bodies as a result of the vital activity and post-mortem degradation of animal and plant organisms. Thus, n-alkanes can be used as molecular indicators to assess potential sources and mechanisms of organic matter transformation in water bodies [15].

Despite many years of monitoring studies of PHC concentrations in the Streletskaya Bay bottom sediments, the composition of AHC, which is a source of information about the progress of the processes of migration, transformation, and accumulation of organic substances in this water area, has not been previously studied. The works carried out to determine the content of certain classes of organic compounds related only to the surface layer of sediments (0–5 cm), and the data of the latest publications on the specified water area are limited to 2015. In addition, the presented works did not study at all the features of the vertical HC distribution in the sediments, their relationship with the geochemical characteristics of bottom sediments, and probable routes of entry, which is a source of information about the chronology of the development of the processes under study.

The purpose of the study is to identify the features of the HC spatial and vertical distribution in BS and to assess the likely sources of their entry into the port coastal water area (using the example of the Streletskaya Bay).

Characteristics of the study area

The Streletskaya Bay is located at the northern coast of the Heracles Peninsula (Fig. 1). Its length is 2.2 km, maximum width – 630 m, width at the exit – 420 m, depth at the bay entrance – 20 m, depth at the top of the bay – 2 m [16]. The shores of the bay are high, with rocky shallows.

According to data [17], the Streletskaya Bay water area receives untreated wastewater from two domestic outlets. In accordance with the storm sewerage scheme of Sevastopol (<https://docs.cntd.ru>), two such lines pass in the immediate vicinity of the bay (Stepanyana Street and Gagarina Avenue). The starting and ending points of the sewers are not indicated, but their waters flow into the bay area. According to some data (<https://sevastopol.su>, <https://primechaniya.ru>), cottage houses are not connected to the central sewerage network. A boat pier, the 91st ship repair plant are located on the territory of the bay, which is densely built up with cottage and apartment buildings. The anthropogenic load on the bay has increased in the last decade as a result of massive development (often without any central sewerage system), increased vehicular traffic along the shore of the bay and the development of small-sized shipping.

¹ AMAP, 2010. Chapter 4: Sources, Inputs and Concentrations of Petroleum Hydrocarbons, Polycyclic Aromatic Hydrocarbons, and Other Contaminants Related to Oil and Gas Activities in the Arctic. In: AMAP, 2010. *Assessment 2007: Oil and Gas Activities in the Arctic – Effects and Potential Effects. Volume 2*. Oslo: AMAP, pp. 4_1–4_85.

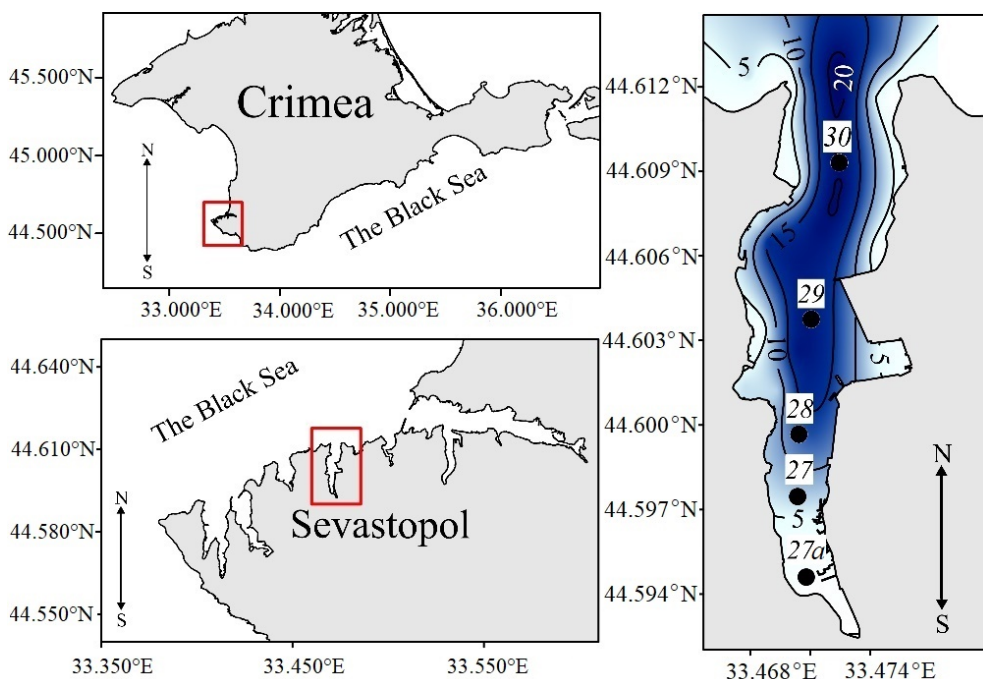


Fig. 1. Schematic map of the bottom sediment sampling stations in the Streletskaia Bay (Sevastopol water area) in summer 2021

Material and research methods

Bottom sediment samples were taken in the summer of 2021 using the Peterson grab with a capture area of 0.038 m² according to a previously approved station scheme (during a joint expedition of the Department of Marine Sanitary Hydrobiology of FRC IBSS and Marine Biogeochemistry Department of FRC MHI as part of long-term monitoring of the Sevastopol bays; station numbering is preserved, Fig. 1). Bottom sediments were placed in sealed bags, labeled, and delivered to the laboratory in refrigeration equipment.

To study the vertical distribution of HC, bottom sediments were collected using a plexiglass tube with a vacuum seal. In the laboratory, the soil column was divided into 2 cm layers using an extruder and an acrylic ring. Subsequently, the sediments were dried to an air-dry state, ground in a mortar, and part of the sample was sifted through sieves with a cell diameter of 0.25 mm.

The granulometric composition of bottom sediments was determined by a combined method of decantation and sieving. The aleurite-pelitic fraction (≤ 0.05 mm) was separated by wet sieving followed by gravimetric determination of dry mass. Coarse-grained fractions (> 0.05 mm) were separated by the sieve method of dry sifting using standard sieves (GOST 12536-2014). C_{org} and C_{carb} content was determined coulometrically on an AN-7529 express analyzer using a method adapted for sea bottom sediments [18, 19]. The standard deviation for samples with C_{org} content $< 0.5\%$ was 0.03%, with C_{org} content $> 1.5\%$ – 0.08%; such a deviation for samples with a C_{carb} content $< 0.6\%$ was 0.07%, for samples with a C_{carb} content $> 8\%$ – 0.09% [19].

The determination of the HC and n-alkanes concentrations was carried out on the basis of the Spectrometry and Chromatography Shared Use Center of FRC IBSS. A dried and sieved sample of 1–2 g was extracted into 150 ml of n-hexane in a Soxhlet apparatus for one hour. The resulting extract was purified on a glass column filled with alumina to remove polar compounds. The resulting extract was concentrated to 1 ml.

An aliquot of the concentrated extract was injected with a microsyringe into the evaporator of a Kristall 5000.2 gas chromatograph with a flame ionization detector (FID) heated to 250 °C. HC separation was carried out on TR-1MS capillary column with a length of 30 m, a diameter of 0.32 mm, and a stationary phase thickness of 0.25 µm (Thermo Scientific). The column temperature was programmed within the range of 70–280 °C (temperature rise rate 8 °C/min). The carrier gas flow (nitrogen) in the column was 2.5 ml/min without flow division. Detector temperature was 320 °C.

Quantitative determination of the total HC content was carried out by absolute FID calibration with an HC mixture, which was prepared by the gravimetric method, with a content within the range of 0.1–5.0 mg/L. The standard sample ASTM D2887 Reference Gas Oil (Supelco, USA) was applied as an HC mixture. The total HC content was determined by the sum of the peak areas of eluted n-alkanes and the unresolved background (Unresolved complex mixture – *UCM*). To process the results, we used Chromatek Analyst 3.0 software, the method of absolute calibration and percentage normalization.

Identification of n-alkanes was carried out using a standard sample of a mixture of paraffin hydrocarbons in hexane with a mass concentration of each component of 200 µg/ml, pristane + phytane – 100 µg/ml in hexane (Supelco, USA).

Not all HC entering BS can be distinguished by gas chromatography [20], for example, cycloalkanes (naphthenes). A mixture of this kind of organic substances is called *UCM*, or a “hump.” During the transformation of organic substances, including those of petroleum origin, more stable and complex compounds are often formed in BS, therefore, the presence of *UCM* in the chromatogram often indicates the presence of degraded HC in BS. It should also be taken into account that the configuration of *UCM* depends on its composition. The nature of the “hump” of natural and anthropogenic HC differs [21]. Anthropogenic compounds are characterized by the “hump” in the high molecular weight region. The maximum in the low-molecular region occurs due to microbial degradation of natural organic compounds, in particular plant detritus [22].

Unresolved complex mixture was calculated by subtracting the peak areas of chromatographically separated HC from the total area of the chromatogram.

In order to analyze the origin and pathways of HC entering bottom sediments, the markers were used, i.e. ratios of certain n-alkanes or their amounts. In this work, attention was paid to the following indices and ratios: *UCM/n-alkanes*, *LWH/HWH*, *P_{aq}*, *TAR*, *ACL*, *TMD*, *C₃₁/C₁₉*, *CPI₂*, *C₃₁/C₂₉* (detailed spelling of abbreviations is given below).

The *UCM/n-alkanes* ratio is the ratio of the chromatographically unresolved background to the n-alkanes content. If the value of the *UCM/n-alkanes* marker is > 10, then it is possible to judge about chronic pollution of

BS. If it is below this indicator, then an active modern supply of petroleum products can be assumed [23, 24].

The *LWH/HWH* ratio (Low-molecular Weight to High-molecular Weight Homologies ratio) reveals the ratio of low-molecular to high-molecular alkanes [25, 26]:

$$LWH/HWH = \Sigma(C_{13} - C_{21})/\Sigma(C_{22} - C_{37}). \quad (1)$$

If high-molecular weight n-alkanes (*HWH*) dominate, the index value will be < 1, which means the dominance of degraded petroleum products containing high-molecular weight HC in their composition. If the predominance of low-molecular weight n-alkanes (*LWH*) is noted, then the index value will be > 1, which means the entry of fresh petroleum products into BS or biogenic autochthonous HC.

The P_{aq} index (Proxy for aquatic macrophytes) makes it possible to find out which type of vegetation (terrigenous or aquatic) predominates in the process of organic matter formation in BS [27]:

$$P_{aq} = (C_{23} + C_{25})/(C_{23} + C_{25} + C_{29} + C_{31}). \quad (2)$$

The *TAR* (Terrigenous/aquatic ratio) index is the ratio of terrigenous matter (allochthonous) to autochthonous, which determines the ratio of terrigenous vegetation to algae in BS [28]:

$$TAR = (C_{27} + C_{29} + C_{31})/(C_{15} + C_{17} + C_{19}). \quad (3)$$

It is believed that the predominance of odd high-molecular weight n-alkanes is observed in the epicuticular wax of higher terrestrial plants, while low-molecular weight odd n-alkanes appear in BS as a result of the vital activity of algae and cyanobacteria [29, 30]. This index provides the determination of not only the type of vegetation that predominates in BS, but also the main source of material entering BS – allochthonous or autochthonous.

ACL index (Average chain length) represents average chain length. It is based on the average quantity of odd carbon numbers of n-alkanes of higher plants [31]:

$$ACL = ((27 C_{27} + 29 C_{29} + 31 C_{31} + 33 C_{33} + 35 C_{35} + 37 C_{37}) / (C_{27} + C_{29} + C_{31} + C_{33} + C_{35} + C_{37})). \quad (4)$$

This index is applied to identify changes in the ecosystem. It remains stable for a long time and decreases sharply when oil pollution occurs. In warm climate, plants produce longer chain n-alkanes; forest plants have a shorter chain length than steppe ones [32–34].

The *TMD* (Terrestrial marine discriminant) index shows the ratio of the terrigenous to water component in BS:

$$TMD = (C_{25} + C_{27} + C_{29} + C_{31} + C_{33}) (C_{15} + C_{17} + C_{19} + C_{21} + C_{23}). \quad (5)$$

A value > 1 corresponds to a dominant terrigenous contribution, while a value < 0.5 corresponds to a dominant marine contribution. Values within the range of 0.5–1 are typical for mixed ecosystems such as estuaries.

C_{31}/C_{19} ratio, like the *TMD* index, demonstrates the ratio of the allochthonous component to the autochthonous component [35].

CPI (Carbon preference index) is based on the predominance of odd or even n-alkanes [36]; CPI_2 is the index where only high-molecular weight part is used to calculate the even-odd predominance:

$$CPI_2 = (\Sigma(C_{23}-C_{31})_{\text{odd}} + \Sigma(C_{25}-C_{33})_{\text{odd}}) / 2\Sigma(C_{24}-C_{34})_{\text{even}}. \quad (6)$$

It is considered that if $CPI_2 < 1$, then this indicates a predominantly biogenic origin of HC in BS, but if $CPI_2 \approx 1$, then this means a large proportion of petroleum-origin HC in BS [37]. The presence of high CPI_2 values is also characteristic of the biogenic nature of organic compounds [21].

Index C_{31}/C_{29} is the ratio of herbaceous to woody vegetation [38]. Herbaceous vegetation in this ratio is above-ground vegetation that dies off every autumn, and woody vegetation includes shrubs (for example, reeds) and trees.

For statistical data processing, the Statistica 12 software package was applied. Cluster analysis of sediment layers sampled in the Streletskaya Bay (station 27) was carried out based on the data concerning total HC content, *UCM* size, and compositional features of n-alkanes (based on calculated markers).

Results and discussion

Geochemistry of bottom sediments. Bottom sediments granulometric composition in the water area of the Streletskaya Bay is heterogeneous. At the top (station 27a) and the central part (stations 28, 29) of the studied water area, BS are represented by dark gray liquid silt with a small amount of sand and a characteristic odor of hydrogen sulfide. At the station located further to the sea (station 30), the marine soils are represented by gray silts with an admixture of fine sand. Natural humidity varies from 72% in the apex part to 57% closer to the bay exit. In the surface layer of dark gray silts in the southern part of the bay (station 27), C_{org} content was 6.3%, which is explained by the increased content of the silt fraction with inclusions of rotted organic matter on the sediment surface and weak water exchange in this part of the water area. Towards the exit from the bay, C_{org} content decreases, amounting to minimum for the water area 4.28% at station 30. Thus, average C_{org} content in the surface layer of BS in the Streletskaya Bay (5.13%) is significantly higher compared to other bays of the Sevastopol region [39–41]. For instance, according to the data from [39–41], average C_{org} content in BS was as follows: in the Kamyshovaya Bay – 1.2% dry wt., in the Kruglaya Bay – 1.4% dry wt., in the Balaklava Bay – 1.97% dry wt., in the Kazachya Bay – 2.7% dry wt., in the Sevastopol Bay – 3.7% dry wt.

Features of C_{org} vertical distribution in the Streletskaya Bay BS indicate the accumulation of the organic component in recent years and C_{carb} reduction (Fig. 2). This is the result of active siltation of the bay due to the intensive supply of organic matter with sewage and storm drains, intensive growth and further death of herbaceous vegetation, as well as weak water exchange, especially in the apex part.

BS hydrocarbon composition. The average concentration of HC in the BS surface layer was 1160 mg/kg with a minimum at the exit from the bay (station 30) – 328 mg/kg and a maximum at the top (station 27a) – 2175 mg/kg (Fig. 3, a). A clear trend towards a decrease in the content of the studied parameter from the mouth of the bay to its exit was noted. By contrast, work [42] presents previously recorded HC concentrations in BS for the other Black Sea coastal waters – the Gelendzhik Bay (11–252 mg/kg), the coast of Greater Sochi (5–119 mg/kg), the Feodosia Gulf (17–80 mg/kg). Thus, it is possible to judge of a fairly high level of BS hydrocarbon pollution in the water area under study compared to other port waters of the Black Sea coast.

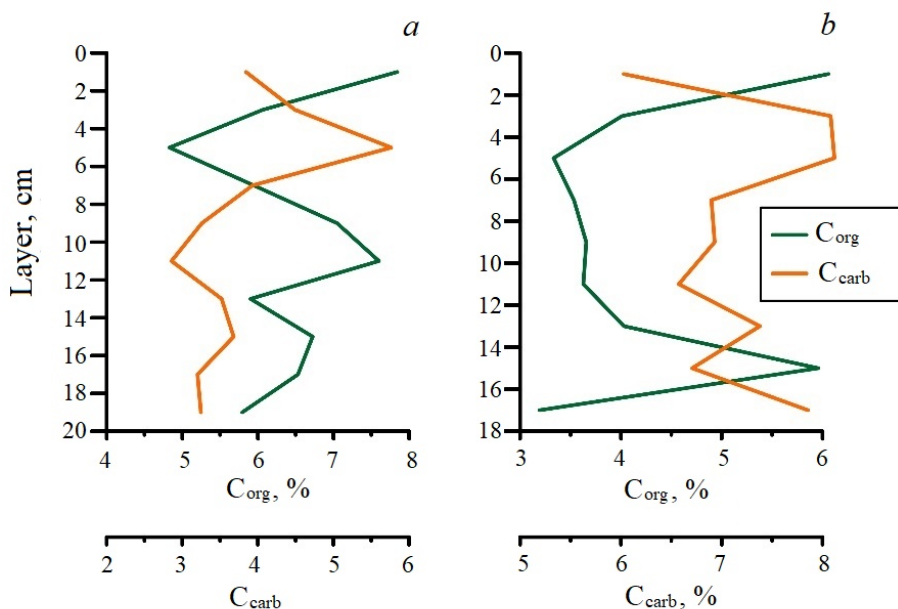


Fig. 2. Vertical distribution of C_{org} and C_{carb} in bottom sediments of the Streletskaia Bay at stations 27 (a) and 30 (b)

UCM was noted in all chromatograms; its content increased gradually from 238.9 mg/kg at the exit from the bay to 1723 mg/kg in its apex part, and the average concentration was 908 mg/kg (Fig. 3, b). The presence of *UCM* indicates a long-term supply of hydrocarbons to BS. It is known that the configuration of the unseparated background depends on the composition of the hydrocarbons entering BS and most often (in coastal areas) is bimodal. The unseparated background curve at the sampling stations was double-humped (an example is given in Fig. 4).

In the BS surface layer in the Streletskaia Bay, it was possible to identify n-alkanes within the range C_{15} – C_{35} (Fig. 5). Compounds with hydrocarbon chain lengths up to C_{33} were present at all sampling stations. The presence of higher molecular weight homologues was typical only for the apex parts of the water area (stations 27a, 27, 28).

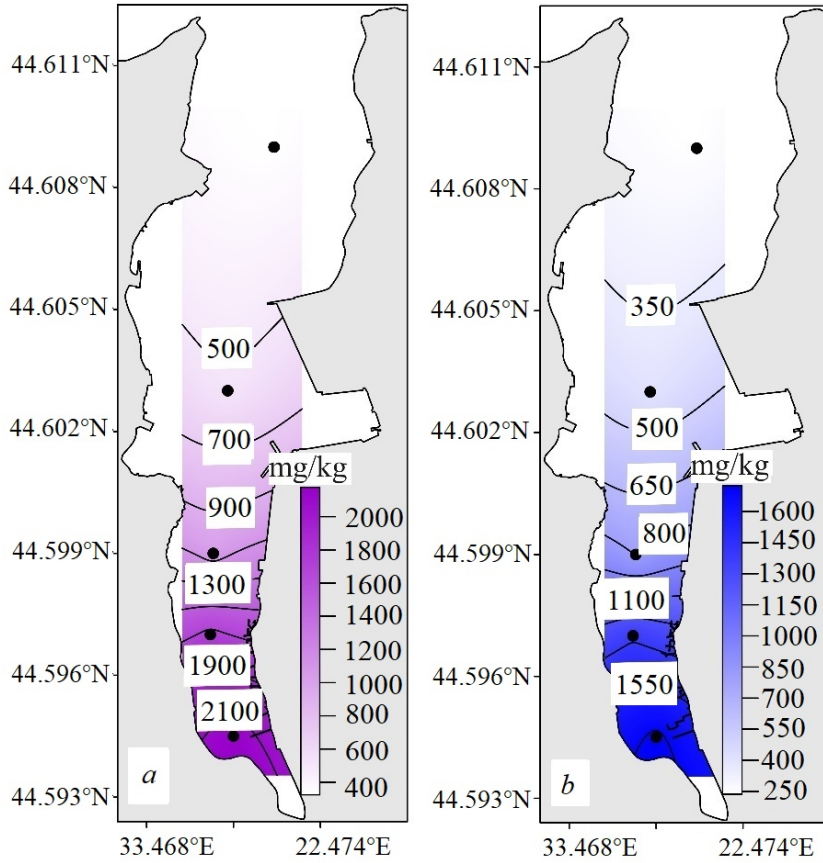


Fig. 3. Spatial distribution of hydrocarbons (a) and UCM (b) in bottom sediments of the Streletskaya Bay

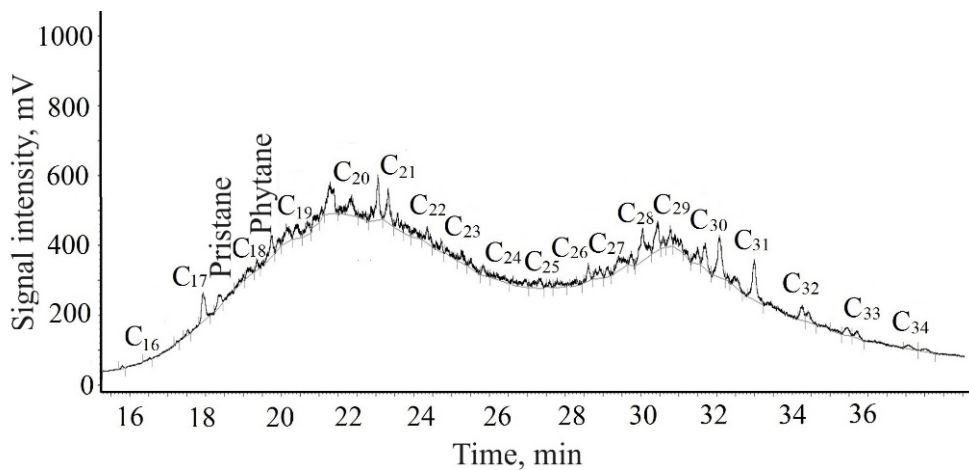


Fig. 4. Example of the n-alkanes chromatogram resulted from the analysis of bottom sediment samples at station 27a

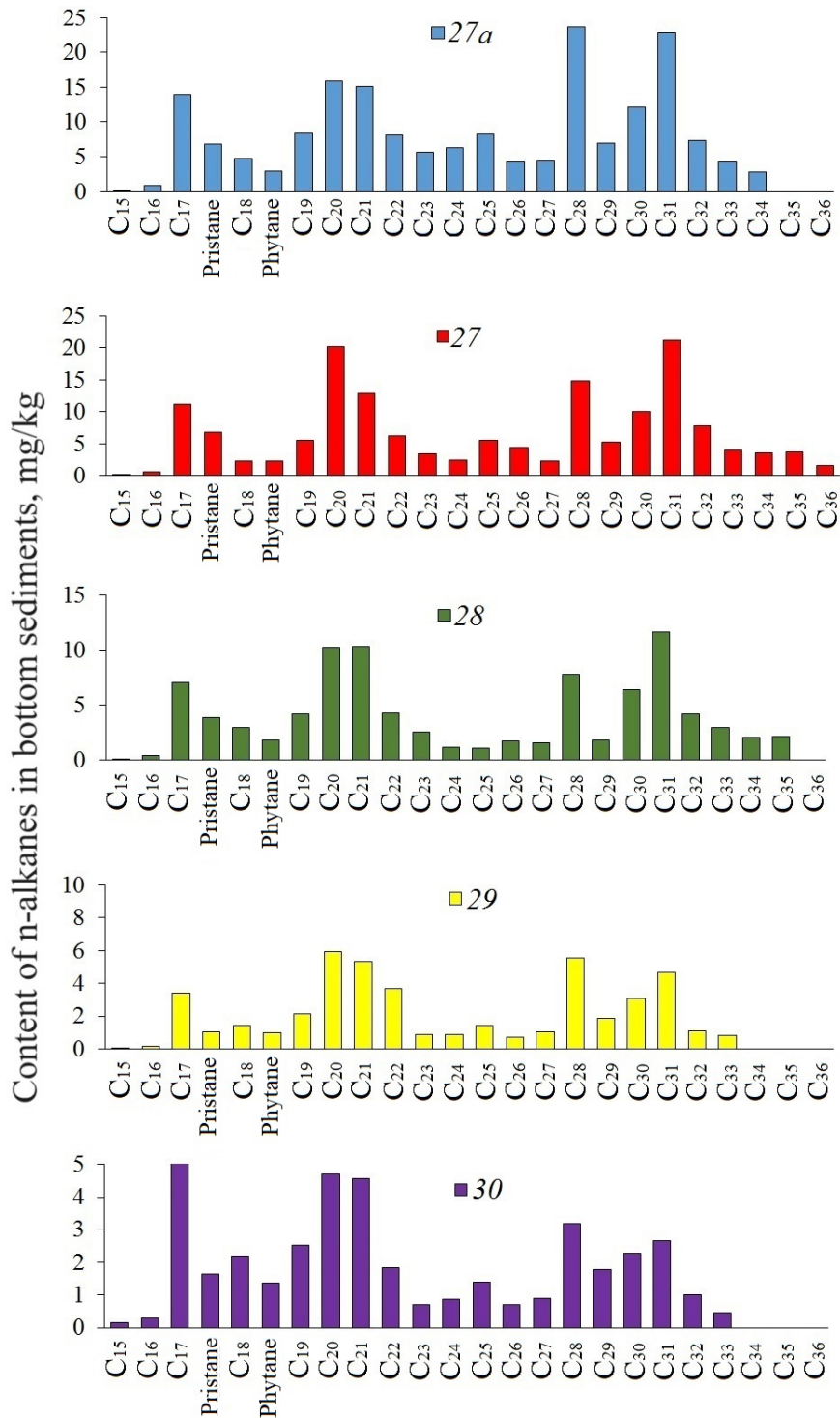


Fig. 5. Composition of the identified n-alkanes in bottom sediments of the Streletskaya Bay at some stations

In all studied parts of the water area, the distribution of homologues was bimodal, which characterizes the duality of HC sources. The first peak occurred in compounds within the range of C₁₉–C₂₂, the second one – mainly within the range of C₂₈–C₃₁. Together with the values of the *CPI* indices (Table), which are close to unity in most cases, this indicates the accumulation of biosynthesis products of phytoplankton organisms in BS and HC microbial destruction, including those of petroleum origin [21, 43]. Moreover, the absence of significant amounts of C₁₆ and C₁₈ homologues can indicate a lesser role of microbial destruction, which results in the accumulation of these compounds [21]. In the high-molecular part of the spectrum, the maximum concentrations were found in homologues within the range C₂₈–C₃₂. Odd compounds in this spectral region are predominantly of terrigenous origin [29]. In this case, attention should be paid to the presence of even homologue C₂₈ in significant quantities, which can indicate the presence of chemical re-synthesis products of organic residues of biogenic (mainly plant) origin, the high content of which underestimates *CPI*₂ values [44].

The calculated diagnostic indices for determining the HC genesis of hydrocarbons in sediments are given in the table. Data from all stations show a predominance of pristane over phytane, which, on the one hand, indicates a larger proportion of biogenic compounds [21], on the other hand – reflects the presence of oxidative conditions in BS [45], promoting the transformation of organic substances.

The *UCM*/n-alkanes marker values were slightly below 10 (average 8.75), which, along with the double-humped form of *UCM*, indicates the presence of transformed compounds of both biogenic and petroleum nature. The *LWH/HWH* index was less than one at all stations, which indicates the predominance of terrigenous material. The exception was station 30, where a slight predominance of petroleum-origin HC was noted. At the same time, a smaller proportion of oil HC in the composition of the hydrocarbon mixture at high levels of HC content in BS and the presence of a significant unseparated background at all sampling stations most likely indicates an intensive HC supply from the shore than the absence of oil pollution. At all stations, *P_{aq}* index fluctuated between the values of 0.1 and 0.3, which corresponds to the predominant contribution of land macrophytes to the BS of the bay.

TAR was > 1 at stations 27a–29, which indicates a predominantly terrigenous supply of material characteristic of coastal areas and seems to be natural for the interior part of the bay. At station 30, it is equal to 0.7; this is a sign of the autochthonous formation of material. The *ACL* index values (28.9–30.5) indicate a mixed contribution of herbaceous and woody vegetation to the bay BS.

TMD index at stations 27a and 27 was > 1, which indicates the predominant contribution of terrestrial vegetation to the organic matter formation in BS. At the remaining sampling stations, the index fluctuated within 0.5–0.8, and this is a sign of the equivalent contribution of terrestrial and marine plants to the formation of the BS hydrocarbon composition.

High values of ratio C₃₁/C₁₉ (> 1) can indicate a dominant contribution from terrestrial vegetation. The index values decrease gradually from the apex part to the exit from the bay, which is in good agreement with the visual determination of

a large number of algae at stations 27a and 27. The index of the ratio of herbaceous vegetation to woody vegetation C_{31}/C_{29} was maximum at station 28, but at other stations it was > 1 . This indicates the predominance of herbaceous vegetation during the formation of the bay BS organic matter, and can also show that oil products have already been transformed.

Values of diagnostic indices for bottom sediments in the Streletskaya Bay

Index	Range of values	Origin	Source
UCM/n -alkanes	6.4–9.8	< 10 – fresh petroleum products > 10 – degraded petroleum products	[23]
LWH/HWH	0.5–1.1	< 1 – higher vegetation, terrigenous ≈ 1 – petroleum, planktonic	[26]
P_{aq}	0.2–0.3	0.1–0.4 – terrigenous	[27]
TAR	0.7–1.7	< 1 – predominance of aquatic vegetation > 1 – predominance of terrigenous vegetation	[46]
ACL	28.9–30.5	mixed contribution of herbaceous and woody vegetation	[47]
TMD	0.5–1.2	$0.5 < TMD < 1$ – mixed contribution of land and sea sources (stations 28–30) > 1 – dominant contribution of allochthonous compounds (stations 27a, 27)	[48]
C_{31}/C_{19}	1.1–3.8	predominance of terrigenous matter	[49]
CPI_2	0.9–1.0	≈ 1 – petroleum alkanes	[21, 50]
C_{31}/C_{29}	1.5–6.3	predominance of woody vegetation	[31]
Pr/Pn	1.2–3.0	< 1 – petrogenic origin $1 < Pr/Pn < 3$ – signs of oxidizing conditions of sedimentation > 1 – biogenic origin	[44, 51, 52]

Thus, most of the markers indicate the active supply of allochthonous substances into BS, which is typical for coastal waters, especially enclosed ones [7], and is also associated with the greater resistance of allochthonous compounds to biodegradation, leading to their disposal in bottom sediments². At the same time, the input and accumulation of petroleum-origin compounds and autochthonous material into BS were recorded. It can be concluded that the origin of HC in the Streletskaya Bay BS is mixed.

High positive correlation was noted between the HC distribution in the surface layer of bottom sediments and C_{org} content (0.96–0.98). The maximum values of correlation coefficients with C_{org} were noted for TMD (0.77), CPI_2 (0.83), and C_{31}/C_{29} (0.87). This confirms the well-known fact [53] that in finely dispersed sediments characterized by high C_{org} content, the accumulation of substances, including those of organic origin, occurs more actively.

² Vykhristyuk, L.A., 1980. [Organic Matter of Lake Baikal Bottom Sediments]. Novosibirsk: Nauka, 80 p. (in Russian).

According to [54], the rate of the Streletskaya Bay sedimentation is 3.5 mm/year. Thus, the selected BS layer corresponds to ~ 65 years, and every 2 cm – to 6 years. This makes it possible to analyze the history of HC accumulation over half a century of human activity and assess the anthropogenic load on the water area (Fig. 6). The BS column was collected in the most polluted apex part of the bay (station 27).

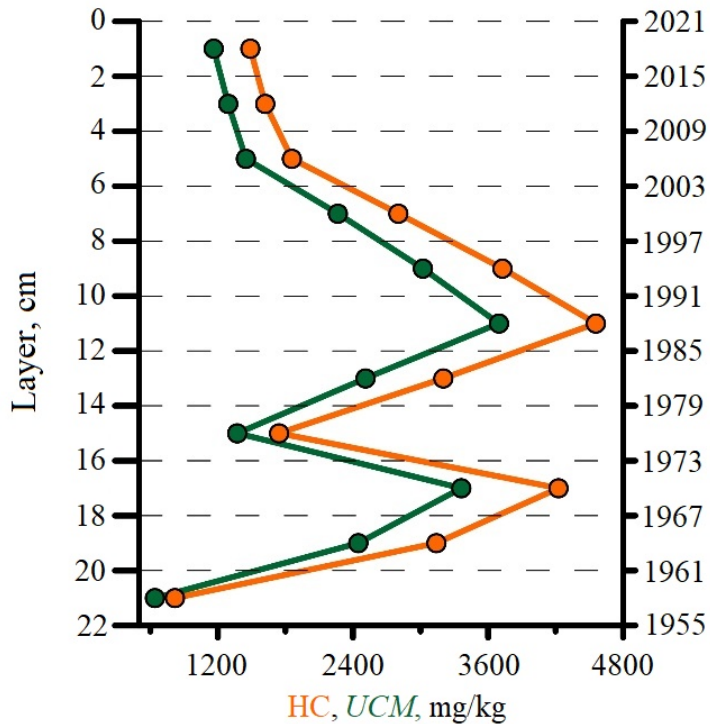


Fig. 6. Vertical distribution of hydrocarbons and *UCM* in bottom sediments of the Streletskaya Bay (station 27)

A change in the content of HC and *UCM* along the horizons was noted. In the layers corresponding to the modern period (0–2, 2–4 cm), the concentration of hydrocarbons and *UCM* is constant, which demonstrates a stable rate of HC entry, transformation, and accumulation in the water area BS. Then HC concentrations increase with depth, from 1488 mg/kg (layer 0–2 cm) to 4558 mg/kg (layer 10–12 cm). As we move deeper into BS, the HC concentration decreases to 3202 mg/kg in layer 12–14 cm. One of the minimum values of HC concentration (1743 mg/kg) was noted at a depth of 14–16 cm, followed by a sharp increase (layer 16–18 cm) and a depression in layer 18–22 cm (Fig. 6).

Analysis of the history of AHC accumulation showed that maximum concentrations in BS are typical for the periods 1967–1973 (layer 16–18 cm) and 1985–1991 (layer 10–12 cm). The accumulation of *UCM* in BS occurred synchronously with the accumulation of HC.

In order to identify periods with different anthropogenic load on the Streletskaya Bay water area, clustering of BS horizons (Fig. 7) was carried out in accordance with the total HC content, *UCM* sizes, and compositional features of n-alkanes (based on the markers). Two groups of horizons with significant differences were identified. The first large cluster includes horizons 6–14 and 16–20 cm, which correspond to the periods 1979–2003 and 1961–1973. These periods included years with high anthropogenic load on the water area. The second group of layers, according to cluster analysis, is represented by the horizons with layer depths of 0–6 and 14–16 cm, which, judging by the given BS dating, correspond to the periods 2003–2021 and 1973–1979.

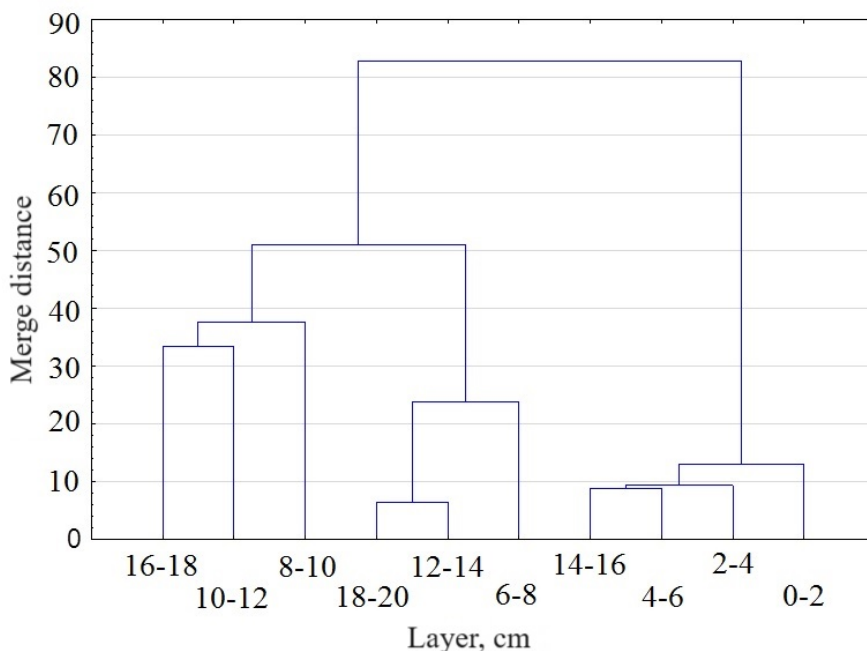


Fig. 7. Results of cluster analysis of the sediment layers sampled in the Streletskaya Bay (station 27)

Differences in the number of HC in BS can be associated with the history of the region development, periods of industrialization, and growth of urban areas. In the 1960s–1970s, construction in the Streletskaya Bay area started, and a residential microdistrict began to form on its left bank. From 1991 to the present, the HC concentrations in the bay bottom sediments have been decreasing, which indicates a decrease in anthropogenic load on the water area. This decrease in concentrations can be associated with the decline in economic growth in the USSR, which affected the industrial production of the city, with the further cessation of the bay exploitation by military and commercial vessels, and with the current lack of production expansion near its waters.

To understand the origin of HC in the bay BS, diagnostic indices of n-alkanes were calculated (Fig. 8).

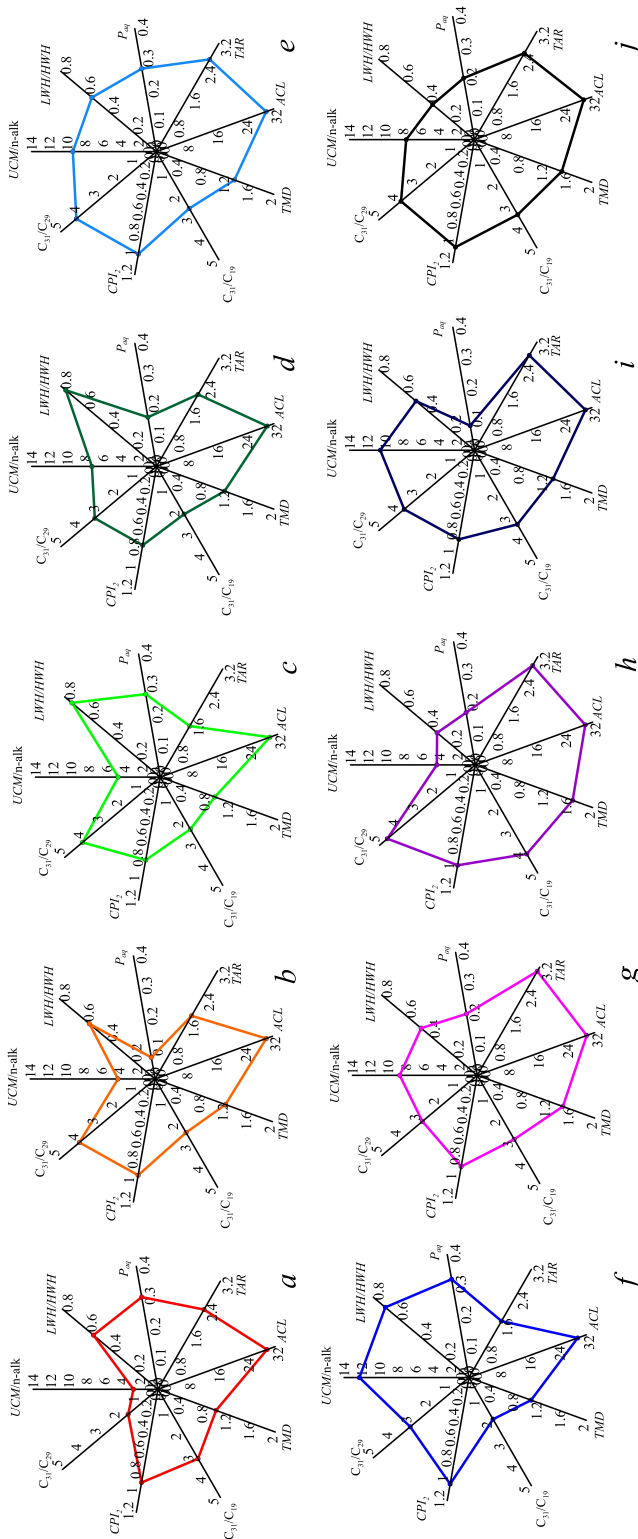


Fig. 8. Distribution of the main markers in the bottom sediments of Stretetskaya Bay (station 27) by layers: (a) 0–2 cm; (b) 2–4 cm; (c) 4–6 cm; (d) 6–8 cm; (e) 8–10 cm; (f) 10–12 cm; (g) 12–14 cm; (h) 14–16 cm; (i) 16–18 cm; (j) 18–20 cm

UCM/n-alkanes ratio varied from 1.76 to 12.29. The maximum values of this marker, ~ 10 and above, were observed at the most polluted horizons (8–12, 16–18 cm); the presence of an increased content of hydrocarbons in BS is associated with the influx of petroleum components into them.

LWH/HWH index values were 0.32–0.76, indicating a predominance of terrigenous material. *TAR* index varied within 1.5–3.1, which indicates the predominance of allochthonous material entering BS. A slight change in the *ACL* index (from 29.7 to 30.3) indicates the stability of HC input processes and the absence of sharp changes in the levels of anthropogenic impact.

C_{31}/C_{29} ratio, in its turn, showed the predominance of herbaceous vegetation in the BS material, which is typical for this area with no forest cover, where coastal landscapes are represented by steppes.

TMD index had values within the range of 0.93–1.49, which indicates different origin of BS matter: for horizons 0–2 and 4–6 cm, a mixed contribution of terrestrial and marine plants to BS was noted; for other horizons, a dominant contribution of material from the land was observed. The ratio of C_{31}/C_{19} n-alkanes varied within the range of 1.9–3.5, which indicates the dominance of allochthonous substances over autochthonous ones. CPI_2 index had values close to 1 (0.9–1.0), which, on one hand, can indicate the presence of oil pollution during the study period and, on the other hand, can be associated with the presence of homologue C_{28} in high concentrations (9–17% of the total n-alkanes), which is of natural origin [44].

When interpreting data on the composition of n-alkanes in BS sampled from different depths, it should be taken into account that many markers that differentiate the origin of hydrocarbons are “strictly” applicable only to fresh organic matter [21]. In this work, we consider deeply transformed compounds. At the same time, the calculated values of HC genesis markers are quite understandable for a given water area, taking into account the characteristics of its location and type of use.

Conclusions

BS composition promotes the HC accumulation: aleurite-pelitic silts with high natural humidity are found everywhere, with the exception of the station at the mouth of the bay (station 30). The average content of organic carbon in the surface layer (5.1%) and in the thickness (5.3%) of the Streletskaya Bay BS is much higher than the values obtained in other bays of the Sevastopol region (1.2–3.7%).

The increase in HC concentrations in the surface layer of the Streletskaya Bay BS from its top (328 mg/kg) to the apex part (2175 mg/kg) corresponds to the patterns of distribution of organic substances identified in previous years and is a consequence of processes in a semi-enclosed anthropogenically loaded water area, namely, difficult water exchange in the bay; distribution of untreated sewage sources along its coast, domestic and storm water rich in organic matter; vital activity of hydrobionts and benthic organisms.

According to the estimates obtained, the Streletskaya Bay BS contain a significant amount of HC, their levels correspond to and sometimes exceed those in the polluted port waters. Nevertheless, the composition of n-alkanes and the nature of the chromatograms indicate a mixed origin of hydrocarbons with a predominance of

allochthonous (coming from land) compounds, as well as the presence of n-alkanes of autochthonous and petroleum origin.

According to the data obtained, the high anthropogenic load on the bay occurred in the periods 1967–1973 and 1985–1991, when the city was intensively developing and the population of the microdistrict adjacent to the Streletskaya Bay was growing. In recent years, the intensity of HC accumulation has been decreasing, while the hydrocarbon background remains high.

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Konstantin I. Gurov – formulation of goals and objectives of the study; presentation of data in the text and their analysis; paper correction; discussion of the study results; preparation of graphic and text materials; formulation of the conclusions

Olga V. Soloveva – analysis of literature data; participation in the discussion of the paper materials; editing and supplementing the text of the paper; paper correction; advisory assistance

The authors have read and approved the final manuscript.

The authors declare that they have no conflict of interest.