


Original article

Geochemical Characteristics and Features of Spatial Distribution of Heavy Metals and Microelements in the Sea of Azov Bottom Sediments

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Abstract

Purpose. The purpose of the study is to assess the geochemical characteristics (granulometric composition, content of organic and inorganic/carbonate carbon) and their impact on the features of spatial distribution of heavy metals in the surface layer of bottom sediments in the southern and central Sea of Azov.

Methods and Results. The data obtained during the expeditions at the R/V *Professor Vodyanitsky* in 2019–2020 were used. The sediments surface layer (0–5 cm) was sampled by the Peterson grab sampler (capture area is 0.1 m²). The analysis of a modern data set on the sediments granulometric composition of the bottom surface layer in the Azov Sea made it possible to assess the patterns in spatial distribution of the following fractions: gravel (10–1 mm), sand (1–0.1 mm), aleurite-pelitic (0.1–0.05 mm), and pelite-aleuritic (< 0.05 mm) ones. The presence and degree of anthropogenic pollution of sediments with heavy metals were assessed by calculating various pollution indices (CF , I_{geo} , PLI , and C_{deg}) including the values of average characteristic concentration which leveled out the impact of the sediments type. The obtained results showed that the Azov Sea bottom sediments were formed predominantly by pelite-aleuritic silts (on average 77%) with fragmentary inclusions of sandy material (on average 5%), and at some stations, a predominant portion of shell detritus (on average 18%) was noted. The organic carbon content varied from 0.6–1.9% in the gravel-sand sediments to 2.6–3.1% in the aleurite-pelitic silt sediments with an average value 2.0% ($n = 15$). Among the trace elements determined in pore waters, the reduced forms of iron and manganese were predominant. It was noted that, on average, all the determined trace elements (except for Sr and Mn) had shown, on the one hand, a negative correlation with the gravel and sand fractions, and with the carbonate carbon content, and, on the other hand, a positive correlation with the portion of clay material, the pelitic fraction contribution, and the organic carbon content.

Conclusions. Application of modern *in situ* data on the geochemical characteristics of bottom sediments made it possible to confirm that the main factors in the sedimentation process in the Sea of Azov are the coastal abrasion, the biogenic sedimentation and the bottom topography features. The resulting assessments of content of trace elements in the bottom sediments characterize them as predominantly unpolluted and moderately polluted. Exceeding of the multiplicity of average characteristic concentration, as well as moderate pollution level (by I_{geo} index) were observed for Cr, Cu, and Zn.

Keywords: Sea of Azov, bottom sediments, granulometric composition, organic carbon, heavy metals, pollution indices, average characteristic concentration, geoaccumulation index

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Introduction

The Sea of Azov is a unique shallow water body with intensive economic use; it has the status of a fishery reservoir of the highest category [1] and plays an important role in shaping the hydrological and hydrochemical regime of the Azov-Black Sea basin

The surface layer of modern bottom sediments of the Sea of Azov is formed under a semi-arid climate in a semi-enclosed basin and is characterized by significant lithological diversity; the main sources of terrigenous material are the river runoff of the Don and Kuban, as well as active abrasion of the coasts and bottom [2]

Studying the level of heavy metal (HM) content is an important scientific task, the relevance of which lies in the fact that, unlike organic pollutants, which are to some extent subject to decomposition in the aquatic environment, HM are highly stable and can persist in the environment for a long time, transforming from one form to another.

The gross content of HM characterizes the overall degree of bottom sediment pollution. This indicator is integral and reflects the potential danger of secondary pollution of the water column and the entire aquatic ecosystem [3]. Entering natural water bodies, HM accumulate in living organisms and can reach high concentrations that have a toxic effect [4–7]. Sources of HM input into marine basins are of natural and anthropogenic origin. Natural sources include river runoff (including dissolved and suspended forms) [8–10], weathering of rocks and soils of the catchment area [11], atmospheric deposition [12], abrasion of coasts and bottom [13], input from pore waters of bottom sediments [14], and water exchange with other areas [15]. Considering the shallowness of the Sea of Azov basin, special interest lies in studying the HM migration processes as a result of bottom sediment resuspension during surge and set-up events, as well as wind-wave mixing [16]. Against the background of general natural conditions, anthropogenic anomalies are localized and are mainly associated with port activities [9, 17], untreated stormwater and sewage discharges [18, 19], input from mining and metallurgical [20], as well as agricultural enterprises [21, 22].

The patterns of HM distribution and accumulation in bottom sediments depend on the texture of sediments, their mineralogical composition, redox conditions, desorption processes, and physical transport. A key factor controlling the accumulation and retention of pollutants, including HM, is the granulometric composition¹. It is with the granulometric composition that the mechanical retention of suspended and some colloidal particles by sediments is functionally associated [23]. The results of early studies of these characteristics for the coastal areas

¹ Mitropolsky, A.Yu., Bezborodov, A.A. and Ovsyany, E.I., 1982. *Geochemistry of the Black Sea*. Kyiv: Naukova Dumka, 144 p. (in Russian).

of the Sea of Azov are reflected in works ² [8]. Currently, the geochemical characteristics of bottom sediments are studied mainly to understand the processes of accumulation and spatial distribution of pollutants [24–27].

The results of studies of the Sea of Azov bottom sediments obtained in the last decade indicate that the spatial distribution of pollutants has a pronounced zonality associated both with natural factors and with the features of economic activity in coastal regions [5, 24, 28–32]. At the same time, as noted in [28], bottom sediments play a key role in the accumulation of pollution, acting as an indicator of anthropogenic impact. Particular attention is paid to Taganrog Bay, where, according to [29–31], stable zones of HM accumulation in bottom sediments have formed. The monograph [24] presents the results of studying the migration and transformation processes of HM in various components of the Sea of Azov marine landscapes. These results serve as a basis for understanding the sources of input, as well as the mechanisms of HM distribution and transformation in the sea ecosystem.

Work [25] provides averaged data on the HM content in bottom sediments for 1986–2017 period. It was noted that the range of average annual values for most of the studied metals and arsenic was 1.2–1.7 times, for cadmium and iron – 2.9 times, for chromium – 4.2 times. It was found that elevated concentrations of iron, manganese, chromium, arsenic, and nickel were observed in 1991–1998, and of vanadium, strontium, zinc, chromium, and lead – in 2013–2016.

Furthermore, ecological state of the Sea of Azov ecosystem is examined in a number of publications that study the HM accumulation in hydrobionts and their impact on the fishery productivity of the basin [25]. A comparative analysis of results obtained in [31] with later studies suggests a certain stabilization of the pollution level, although the problem remains extremely relevant. Modern works, for example [5], focus on studying the pollution level and assessing HM fluxes in Taganrog Bay, which opens up new possibilities for the ecosystem state forecasting.

Thus, it is evident that the problem of the Sea of Azov pollution with heavy metals continues to be relevant and requires not only further monitoring but also a detailed study of pollutant transformation processes, which is especially important for developing measures to preserve ecosystem stability and the environmental safety of the region. However, most studies focus on the eastern and northeastern parts of the sea, while bottom sediments in the southwestern part adjacent to the Crimean Peninsula have hardly been studied [26, 32]. It is also extremely important to study the features of HM accumulation and distribution, taking into account their fractional composition and organic matter accumulation. This will allow determining the conditions of development, forecasting changes in the ecological state of the marine environment, and preventing environmentally hazardous phenomena.

Based on this, the study is purposed at assessing the geochemical characteristics and their impact on the HM spatial distribution features in the surface layer of bottom sediments using the example of coastal areas in the southern and central parts of the Sea of Azov.

² Shnyukov, E.F., ed., 1974. *Geology of the Sea of Azov*. Kyiv: Naukova Dumka, 247 p. (in Russian).

Materials and methods

The study applies data obtained during expeditions on the R/V *Professor Vodyanitsky* in 2019–2020. The study area consisted of two sites: the first was located in the southern part of the Sea of Azov adjacent to the Kerch Peninsula, and the second was a transect of four stations in the central part of the sea (Fig. 1).

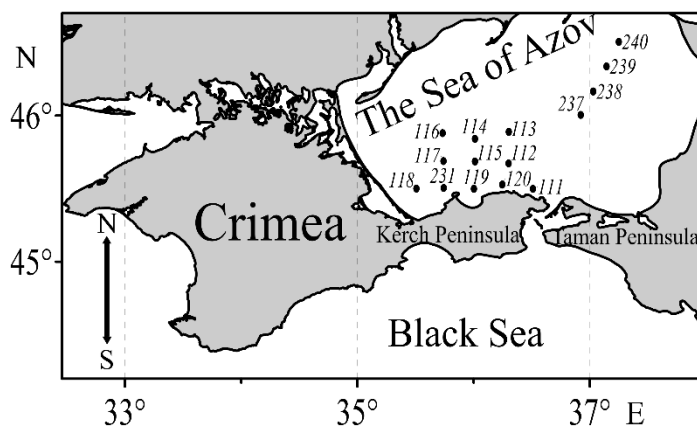


Fig. 1. Location of the Azov-Black Sea region under study and the layout of bottom sediment sampling stations in the Sea of Azov

Surface sediment samples (0–5 cm) were collected using a Peterson grab sampler (capture area 0.1 m²). For subsequent analysis of HM content, the upper 5 cm layer of sediment was taken from the central part of the grab using a plastic spoon. The samples were placed in pre-labeled polyethylene zip-lock bags. Then, in the laboratory, the samples were dried to constant weight at 105 °C followed by homogenization.

Granulometric analysis of bottom sediment samples was carried out by decantation and sieving (GOST 12536-2014). The sieve method was applied to separate and weigh the coarse fractions (> 0.1–0.05 mm). The analysis was performed using a set of sieves with different mesh diameters (10; 7; 5; 2.5; 2; 1; 0.5; 0.25; 0.1; 0.05 mm).

The determination of organic (C_{org}) and carbonate (C_{carb}) carbon content in bottom sediment samples was carried out by dry combustion using an AN-7529M express analyzer according to a method adapted for marine bottom sediments [33]. The root-mean-square deviation (RMSD) for samples with $C_{org} < 0.5\%$ was 0.03%, and for $C_{org} > 1.5\%$ – 0.083%. For C_{carb} , the RMSD varied from 0.07% for samples with $C_{carb} < 1\%$ to 0.09% for $C_{carb} > 8\%$.

To analyze the pore waters of bottom sediments, we applied the polarographic method with a glass Au-Hg microelectrode [34, 35].

To determine the gross content of HM (Fe, Mn, Ti, V, Cr, Ni, Cu, Zn, Sr, Pb, Co) in bottom sediments, the X-ray fluorescence analysis (XRF) method was applied using a Spectroscan Max-GVM spectrometer manufactured by Spectron (Russia)³.

³ NPO Spektron, 2016. *Methodology for Measuring the Mass Fraction of Metals and Metal Oxides in Powder Soil Samples Using X-Ray Fluorescence Analysis M049-P/16*. Saint Petersburg: NPO Spektron LLC, 18 p. (in Russian).

The equipment calibration was performed using a series of certified reference soil samples (typical chernozem, soddy-podzolic sandy loam soil, red soil, and carbonate sierozem). Validation of the calibration curves was carried out using state standard samples DSZU 163.1-98 and DSZU 163.2-98. To assess reproducibility and measurement accuracy, an eight-fold analysis of the certified sample DSZU 16.3.1-98 was performed.

In the absence of regional standards for assessing bottom sediment quality, an approach that levels out the influence of sediment type was applied. It is based on the average characteristic concentration (ACC) of priority pollutants determined in [31] for different types of bottom sediments in the Sea of Azov.

The ratio of absolute concentration to the average characteristic concentration is a dimensionless quantity called the ACC multiplicity [31]

$$\text{ACC multiplicity} = \frac{C_i}{\text{ACC}}$$

where C is concentration of the determined i -th element; ACC is the average characteristic concentration of the i -th element for different sediment types.

If the ACC multiplicity < 1 , then there was practically no fresh input of the determined heavy metals into the given sea area, regardless of the absolute pollution values and the type of analyzed sediment. If the ACC multiplicity > 1 , this area is the one of increased anthropogenic impact in a specific time period.

Average typical concentrations of HM for various types of bottom sediments in the Sea of Azov for 1996–2006 [31]

Soil type	Fe, %	Mn, %	V, mg/kg	Zn, mg/kg	Cr, mg/kg	Cu, mg/kg	Ni, mg/kg	Pb, mg/kg	As, mg/kg
Shell with sand admixture	0.9	0.025	24	17	39	16	25	6	3
Shell with sand and silt admixture	1.6	0.039	47	41	62	25	39	10	5
Silty sand with shell admixture	2.7	0.055	69	79	76	29	51	13	7
Light gray silt	3.7	0.060	84	98	80	35	60	17	8
Gray silt with shells	4.6	0.063	108	111	86	40	69	22	10
Dark dense silt	5.2	0.065	125	120	92	46	75	25	12

The presence and degree of anthropogenic pollution of sediments by HM were assessed by calculating the following parameters: contamination factor (CF) and geoaccumulation index (I_{geo}). When calculating these parameters, which characterize the level of element concentration relative to its background values, the average element concentrations obtained for the bottom sediments of the Sea

of Azov in the period 1991–1995 and presented in [31] were used. These values were: 0.052% for Mn, 3.33% for Fe, 87 mg/kg for V, 38 mg/kg for Zn, 33 mg/kg for Cr, 29 mg/kg for Ni, 23 mg/kg for Cu, 17 mg/kg for Pb, and 11 mg/kg for As.

Integrated pollution indices, which are widely used for the ecological assessment of bottom sediments in the coastal zone, were also determined: pollution load index (*PLI*) and degree of contamination (C_{deg}) [36–39].

The contamination factor was calculated according to [40]:

$$CF = \frac{El_{sam}}{El_{back}},$$

where El_{sam} and El_{back} are the concentration of the element directly in the studied samples (mg/kg) and the background value of the element according to [31]. The contamination factor classes were divided into low ($CF \leq 1$), moderate ($CF = 1-3$), considerable ($CF = 3-6$), and very high ($CF > 6$) [40].

Geoaccumulation index was calculated according to [41]:

$$I_{geo} = \log_2 \left(\frac{El_{sam}}{1.5 \cdot El_{back}} \right).$$

The use of the factor 1.5 is due to the need to account for possible fluctuations in the background values of the element and the negligible contribution of anthropogenic sources [42]. Geoaccumulation index gradation is as follows: $I_{geo} \leq 0$ – practically unpolluted sediments; 0–1 – unpolluted to moderately polluted; 1–2 – moderately polluted; 2–3 – moderately to strongly polluted; 3–4 – strongly polluted; 4–5 – strongly to extremely polluted; $I_{geo} > 5$ – extremely polluted [41].

Pollution load index was calculated according to [43]:

$$PLI = \sqrt[n]{CF_1 \cdot CF_2 \cdot CF_3 \cdot \dots \cdot CF_n},$$

where *PLI* is a value characterizing the integrated pollution index; $CF_{1,2,3...}CF_{1,2,3...}$ are contamination factors for the metals listed above; *n* is a number of metals in a sample (in this work $n = 10$); for polluted sediments $PLI > 1$, for unpolluted sediments $PLI \leq 1$.

The degree of contamination, according to [40], has the form

$$C_{deg} = \sum_{i=1}^n CF_i.$$

A low degree of contamination corresponds to $C_{deg} < 10$, moderate – $C_{deg} = 10-20$, considerable – $C_{deg} = 20-40$, very high – $C_{deg} \geq 40$ [40].

Results and discussion

Geochemical characteristics of bottom sediments

Granulometric composition. The description of the Sea of Azov bottom sediments is widely presented in [44, 45]. It is noted that terrigenous and organogenic bottom sediments are most widespread, while mixed and chemogenic sediments are less represented [44–46]. Based on the bottom topography features and the sea area depth, granulometric composition of bottom sediments varies from sands

of different grain sizes to aleuritic and clayey silts. Numerous bottom topography irregularities, such as banks, spits, and coastal bars, also contribute. Coarse gravel-sand fractions predominantly accumulate here, and there is an increased proportion of authigenic shell material in the sediments [44–46].

Based on the analysis of a modern data set on the granulometric composition of bottom sediments in the Sea of Azov surface layer, the spatial distribution patterns of the following fractions were determined: gravel (10–1 mm; Fig. 2, *a*), sand (1–0.1 mm; Fig. 2, *b*), aleurite-pelitic (0.1–0.05 mm; Fig. 2, *c*), and pelite-aleuritic (< 0.05 mm; Fig. 2, *d*).

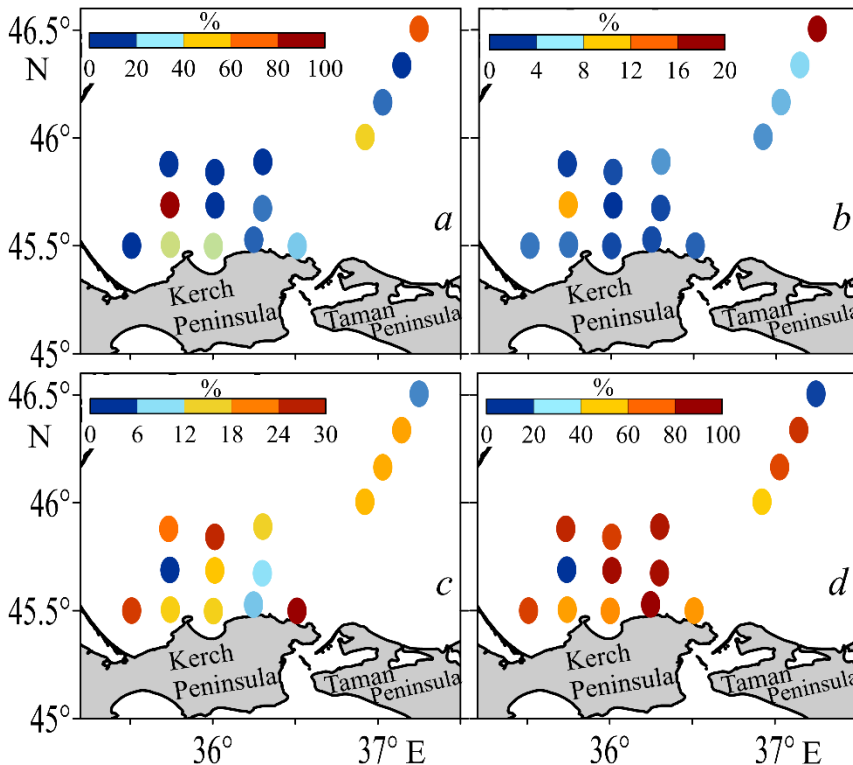


Fig. 2. Spatial distribution of the bottom sediments granulometric fractions: *a* – gravel; *b* – sand; *c* – aleurite-pelitic; *d* – pelite-aleuritic

It was noted that the Sea of Azov bottom sediments are mainly composed of pelite-aleuritic silts. On average, fine-grained material accounts for 76% of all samples, with the pelite-aleuritic fraction accounting for 62% and the aleurite-pelitic fraction for 14%. The increased proportion of gravel-sand material (8% above average) in the central part of the sea area is explained by the abundant inclusion of medium and large shells. Such a local increase in shell material may be a consequence of biogenic sedimentation.

It was also found that an increased proportion of silt material (82–96%), including the pelitic fraction (55–88%), is observed at stations in the Sea of Azov southern part adjacent to the Kerch Strait area. The sand material content varies within the range of 0.2–29% (average 5%); maximum values are noted at st. 117 near

the Kerch Peninsula (17%) and st. 240 in the central part of the sea (29%), and minimum values (0.2–1.2%) in the southern part near the Kerch Strait. In the surface sediment layer, gravel material occurs fragmentarily and consists of medium (5–7 mm) and large (> 10 mm) shells and their fragments. An increased proportion of coarse-grained material (28–80%) is observed at individual stations in the western part of the study area near the Kerch Peninsula, as well as in the central part of the sea (37–65%) (Fig. 2, *a*). The results obtained are consistent with earlier data and the mapping results of the Sea of Azov bottom sediments presented in [46].

The abovementioned distinctive features of bottom sediment distribution in the southeastern (adjacent to the Crimean Peninsula) and central parts of the sea are determined by the bottom topography and sedimentation features of material entering the bottom sediments. Thus, in the central part of the sea at depths of 10–13 m, there is an extensive (area 5 thousand km²) gently undulating accumulative clayey-silty plain (Panov submarine plain), the southern part of which adjoins the Kerch Peninsula. The bottom surface slope is insignificant and faces the Zhelezinskaya Trough [47]. This is consistent with the increase in the proportion of pelitic material from west to east from 53 to 87%. Biogenic processes also play a large role in the formation of the surface layer of bottom sediments. This explains the local maximum of the proportion of gravel-sand material (98%) at st. 117, located at a distance from the shore. Biogenic deposits form the above-water part of accumulative forms, while mineral sands and silts participate in the formation of their underwater slopes [48]. In addition, the configuration and structure of the coastline are subject to storm, seasonal, and multi-year variability [49]. Severe storms contribute to a radical restructuring of coasts and the transport of bottom sediments, significantly expanding the boundaries of the coastal zone. This explains the elevated concentrations of gravel-sand material in the area adjacent to Cape Kazantip in the northern part of the Kerch Peninsula.

The transect located in the central part of the sea partially crosses the Zhelezinskaya Trough (st. 238, 239), and in its northern part (st. 240) – the Achuevskaya Bank. As a result, local maxima of silt material, corresponding to depressions in the relief, and of gravel-sand material, characteristic of underwater accumulative forms (bars and banks), are noted.

The presented results are consistent with the data of earlier studies given in ² [46], and also supplement them with materials obtained in the coastal part of the Crimean Peninsula.

Content of organic and inorganic carbon. Early studies on the distribution and content of C_{org} in the upper layer of the Sea of Azov bottom sediments are presented in [8, 50, 51]. It was noted that the absolute masses of organic carbon in the Sea of Azov sediments are 10 times greater than in the Baltic Sea and 33 times greater than in the White Sea, which is explained by the peculiarities of organic matter mineralization in the water column of colder water bodies and the chemical and bacteriological destruction of organic matter at the water–bottom sediment interface in the southern seas [8, 51]. According to [51], the maximum values (2.64–3.19%) of C_{org} content are characteristic of clayey silts, the minimum (0.26–0.29%) – for sands and coarse silts; concentrations vary depending on the season and also increase from the mouth of the Don River to the central and southern parts of the sea.

Also, this work, taking into account earlier data, determined the features of long-term accumulation of C_{org} in the bottom sediments of the Sea of Azov. It was revealed that these features are cyclic and are determined by the general variability of sedimentation conditions and interannual changes in the hydrological, hydrobiological, and hydrochemical characteristics of the area.

According to the results of the analysis of modern bottom sediment samples from the Sea of Azov, the C_{org} content varies from 0.6–1.9% in gravel-sand sediments to 2.6–3.1% in aleurite-pelitic silts near the Kerch Strait (Fig. 3, *a*) and within the Zhelezinskaya Trough, with an average value of 2.0% ($n = 15$), which is consistent with previously obtained results [51].

At the same time, maximum C_{org} concentrations were observed both for samples with an increased proportion (> 80%) of pelitic material and for sediments with inclusions of gravel-sand fraction (11–30%). Thus, the correlation between C_{org} content and silt fraction was only 0.3, which violates the “classical” understanding of the type of relationship between C_{org} content and granulometric fractions (Fig. 3, *c*).

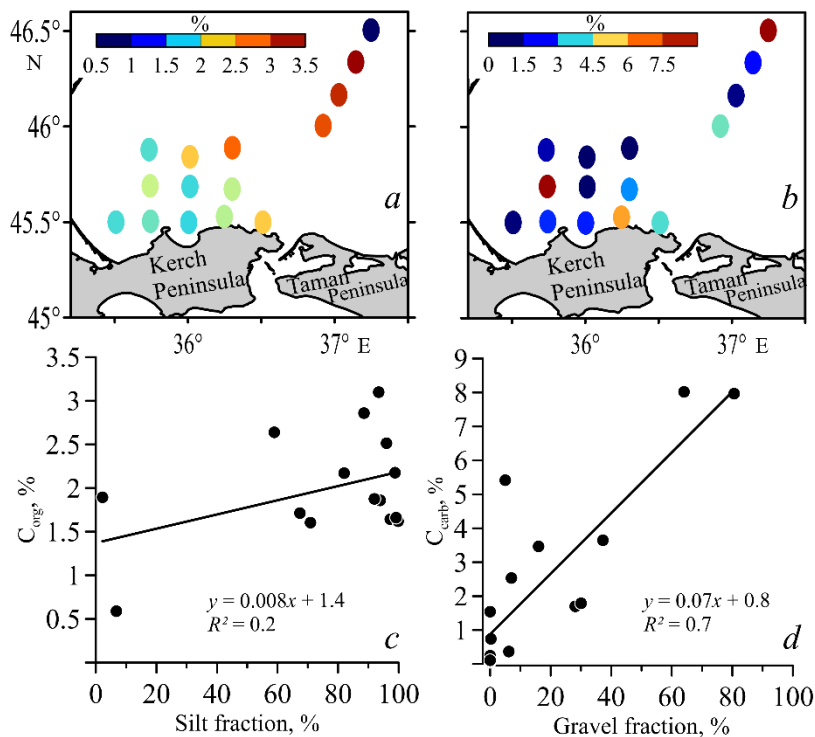


Fig. 3. Spatial distribution of C_{org} (*a*) and C_{carb} (*b*), as well as the relationships between the C_{org} content and the proportions of silt fraction (*c*), and between the C_{carb} content and the proportions of gravel fraction (*d*) in the bottom sediments of the Sea of Azov

C_{carb} content in the surface layer of the Sea of Azov bottom sediments varies from 0.1–0.4% in sediments with a maximum (> 96%) proportion of silt material in the western part of the study area to 4–8% at stations on the transect through the central part of the Sea of Azov (Fig. 3, *b*). The increased C_{carb} content for silty

sediments at st. *111* and *120* is apparently explained by the nature of terrigenous material input as a result of abrasion of the Kerch Peninsula shores. For C_{carb} , the maximum positive correlation was observed for gravel (0.9) and sand (0.8) material, and the minimum negative correlation for silt (-0.9), silty (-0.7), and pelitic (-0.8) material (Fig. 3, *d*).

The revealed spatial heterogeneity of geochemical characteristics of bottom sediments determines the features of the formation of redox conditions in them and regulates the processes of input, accumulation, and redistribution of pollutants.

Redox conditions in bottom sediments. Chemical composition analysis of pore waters of bottom sediments and the features of redox conditions in them was performed for a station located in the southern part of the Sea of Azov at the exit from the Kerch Strait.

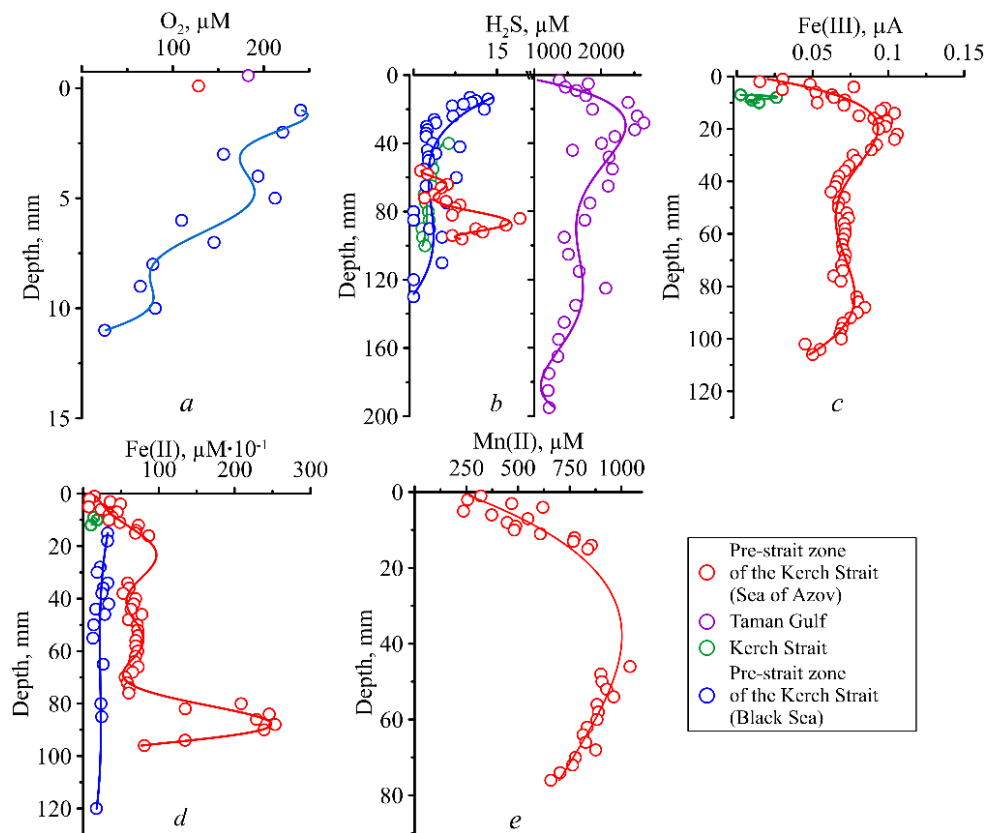


Fig. 4. Vertical distribution of the main components of bottom sediment pore waters for the Sea of Azov and adjacent areas of the Taman Bay, the Kerch Strait and the pre-strait zone from the Black Sea side

High content of organic carbon (2.1%) in fine-grained silts of the upper layer of bottom sediments (99% silt fraction) led to intensive redox processes at the water – bottom interface, as a result of which a sharp oxygen concentration gradient formed and its content decreased to 133 μM at the sediment surface.

The predominant trace components of pore waters were reduced forms of iron and manganese (Fe(II) and Mn(II)). High concentrations of Fe(II) (average 803 μM , maximum 2700 μM) confirm the intensive reduction processes (Fig. 4, *d*). Mn(II) is characterized by a two-layer distribution: in the upper layer (1–15 mm) the concentration increased from 236 to 854 μM , and in the underlying layer (36–76 mm) it decreased from 1042 to 658 μM (Fig. 4, *e*). In the zone where mobile forms of manganese and iron were absent, the presence of iron monosulfide (FeS), a product of Fe(II) interaction with sulfides under sulfate reduction conditions, was recorded. Changes in concentrations with depth are determined by different proportions of fractions: the proportion of gravel-sand material increases with depth from 1.5 to 56%, the proportion of silt decreases from 99 to 44%.

The quantitative characteristics of pore water components at the station in the Sea of Azov differed from those in adjacent areas (Kerch Strait, Taman Bay, Kerch pre-strait zone from the Black Sea side).

Thus, in the pore waters of bottom sediments of the Kerch Strait southwestern part, reduced forms of iron (up to 2769 μM) dominated; Fe(II) content was comparable to its concentration in the most anthropogenically loaded areas of the Sevastopol region [52].

Bottom sediments of Taman Bay are characterized by anaerobic conditions [53], which is confirmed by the complete absence of oxygen and the presence of hydrogen sulfide already at the surface. Its concentration reached a maximum of 2642 μM at a depth of 28 mm (Fig. 4, *b*), and the average concentration (1721 μM) was approximately twice the maximum values characteristic of deep-sea sediments of the Black Sea [54].

Chemical composition of pore waters in the Kerch pre-strait zone from the Black Sea side was characterized by deep (up to 11 mm) penetration of oxygen into the bottom sediments (Fig. 4, *a*), the formation of stable aerobic conditions in the surface layer, as well as the absence of reduced forms of manganese and relatively low Fe(II) concentrations (130–322 μM), significantly inferior to the values at the station in the Sea of Azov. Such characteristics are due to the fact that coarse-grained material predominated in the surface layer of bottom sediments, and the average organic carbon content was 0.7%, which is quite typical for areas with active hydrodynamics (bottom currents in this area reach 20 cm/s).

Heavy metals in bottom sediments. Spatial distribution of the studied HM in the surface (0–5 cm) sediment layer is shown in Fig. 5. The concentrations of elements varied widely: 0.04–0.07% (Mn), 0.14–0.58% (Ti), 1.1–4.9% (Fe), 4–13 mg/kg (As), 0–30 mg/kg (Pb), 0–33 mg/kg (Co), 17–65 mg/kg (Ni), 7–76 mg/kg (Cu), 52–122 mg/kg (Zn), 44–130 mg/kg (Cr), 42–133 mg/kg (V), 128–1253 mg/kg (Sr). Measured values are consistent with modern data obtained for adjacent areas (Kerch Strait [39], Black Sea [55]).

It was found that increased content of titanium (0.58%), iron (4.9%), chromium (130 mg/kg) is noted at stations in the southern part of the Sea of Azov adjacent to the Kerch Strait area. The distribution of trace element concentrations in the bottom sediments of the studied part of the Sea of Azov is heterogeneous. This may be due to the features of bottom geomorphology, hydrodynamic regime, and distribution of geochemical characteristics of bottom sediments. Thus, minimum

concentrations of all studied elements were noted at st. 240, located within the Achuevskaya Bank; the granulometric composition at this station demonstrated an increased proportion of gravel-sand material (93%). The maximum content of arsenic (13 mg/kg), lead (30 mg/kg), and cobalt (33 mg/kg) was determined in the southern part of the studied area. The maximum content of manganese (0.07%), vanadium (133 mg/kg), nickel (65 mg/kg) is observed at stations in the southern part adjacent to the Kerch Strait area and in the Sea of Azov central part. Elevated concentrations of zinc (113–122 mg/kg) are observed throughout the studied area of the Sea of Azov and are noted mainly for sediments with an increased (77–85%) proportion of the pelitic fraction. The maximum strontium content (1253 mg/kg) was determined in the central part of the area under study.

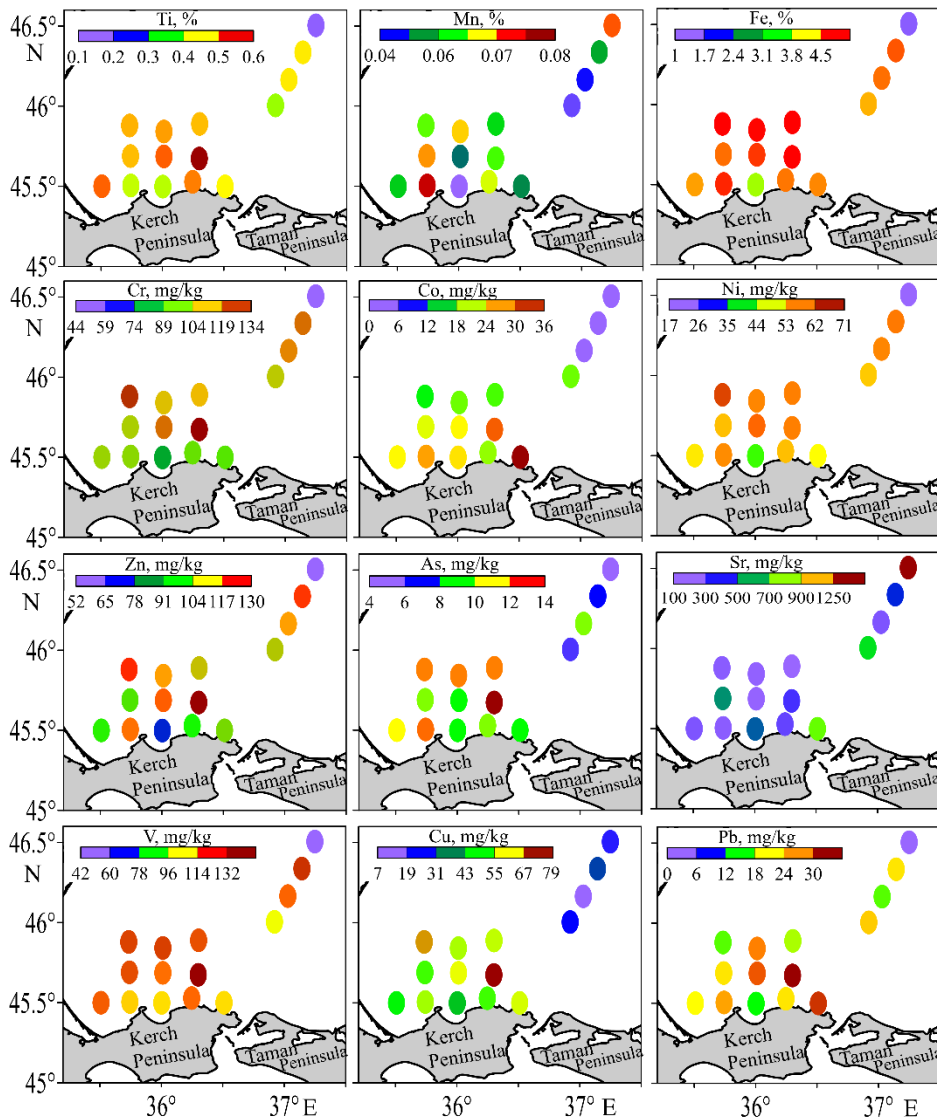


Fig. 5. Spatial distribution of heavy metals and trace elements in the surface layer of bottom sediments of the Sea of Azov

Data comparison with the results of [32] revealed that since 2016, the values for all studied elements have significantly decreased. Additionally, the obtained results were compared with long-term monitoring studies (averaged for 1996 and 2006) published in [31]. It was noted that the change in concentrations for lead, iron, nickel, and arsenic is minimal. For copper, concentrations increased by 12 mg/kg, for zinc – by 14 mg/kg, for chromium – by 25 mg/kg, for vanadium – by 29 mg/kg. The obtained maximum concentrations of Zn and Cu are also consistent with earlier data on the content of these elements in the Sea of Azov bottom sediments presented in [28].

Average ACC multiplicity values for all elements except Mn (0.98) and Ni (0.88) exceed unity, which indicates an increase in the content of the studied elements. Maximum values are noted at st. 117, which is explained by elevated concentrations of elements in gravel-sand sediments. For chromium (1.3) and copper (1.4), the excess of ACC multiplicity is observed at most stations, except st. 237–239 for Cu and st. 240 for Cr.

CF values demonstrated a low pollution level of surface sediments for As (0.8) and Sr (0.8) and a moderate level of pollution for Mn, Pb, Fe (1.1), V (1.3), Ni (1.8), Cu (2.0), Zn (2.6), and Cr (3.0).

According to I_{geo} index gradation, the Sea of Azov bottom sediments are practically unpolluted with Sr (–1.3), As (–1.0), Mn and Fe (–0.5), Pb (–0.4), V (–0.3). In terms of accumulation level of Ni (0.2), Cu (0.2), Zn (0.8), and Cr (1.0), the bottom sediments can be considered unpolluted to moderately polluted. At individual stations, bottom sediments can be considered moderately to strongly polluted: at st. 112 – Zn, Cr, Cu; at st. 113–116, 237–239 – Cr; at st. 240 – Cu.

The values of integrated pollution indices PLI and C_{deg} were 1.39 and 15.48, respectively, which characterizes the Sea of Azov sediments as moderately polluted.

The relationship between the spatial distribution of trace elements and geochemical characteristics of bottom sediments, such as granulometric composition, C_{org} , C_{carb} , was studied. Analysis of the obtained data revealed how groups of trace elements correlate differently with individual geochemical characteristics, which determines the features of their distribution.

On average, for all trace elements (except Sr and Mn), a negative correlation is noted with the gravel and sand fractions, as well as with carbonate carbon content, and a positive correlation with the total proportion of silt material, as well as with the contribution of the pelitic fraction and organic carbon content.

It was found that the maximum positive correlations for Ti (0.7), V (0.7), Cr (0.7), Fe (0.7), Ni (0.7), Zn (0.6), and As (0.5) were noted with the proportion of the pelitic fraction of sediments. For Pb, the maximum positive correlation (0.5) was observed with the proportion of the silt fraction; for Mn (0.4) and Sr (0.8) – with the proportion of sand material. For Co and Cu, the maximum relationships were noted with sand content and were negative (–0.5).

Conclusion

Based on the analysis of modern data for 2019–2020 period, the geochemical features of bottom sediments and chemical composition of pore waters were studied, and quantitative estimates of spatial distribution and pollution level of heavy metals in the Sea of Azov bottom sediments were obtained.

It was found that, in terms of granulometric composition, the bottom sediments in the area of the Panov accumulative plain were represented mainly by pelite-aleuritic silts, consisting of 65% pelite-aleuritic and 15% aleurite-pelitic material. The presence of local areas with an increased proportion of gravel-sand material in the southern part is explained by the features of biogenic sedimentation, storm transport of sediments at stations located near the shore, and in the central part – by the location of the station within the Achuevskaya Bank.

High intensity of organic matter accumulation in the Sea of Azov bottom sediments determined the increased C_{org} content in them. Maximum C_{org} concentrations were observed both for samples with an increased proportion (> 80%) of pelitic material and for sediments with inclusions of gravel-sand material (11–30%). C_{carb} content varied from 0.1–0.4% for silty sediments with minimal inclusion of gravel-sand material to 4–8% for sediments with an increased proportion of shell gravel and detritus, as well as at stations located near the abrasive shores of the Kerch Peninsula.

Quantitative characteristics of pore water components at the station in the Sea of Azov differed from those in adjacent areas (Kerch Strait, Taman Bay, pre-strait zone from the Black Sea side). Intensive redox processes in fine-grained sediments with increased (2.1%) C_{org} content led to the formation of a pronounced oxygen concentration gradient and a decrease in its concentration at the sediment surface to 133 μM . The predominant trace components of pore waters at the station in the Sea of Azov were reduced forms of iron and manganese.

Estimates of the bottom sediment pollution degree were obtained both taking into account differences in their granulometric composition (average characteristic concentration) and using geochemical indices (CF , I_{geo} , PLI , C_{deg}). It was noted that for most of the studied metals, a low and moderate pollution level was observed, not exceeding background values for the Sea of Azov. Exceeding the multiplicity of the average characteristic concentration and an average pollution level (according to I_{geo} index) were observed for Cr (st. 112–116, 237–239), Zn (st. 112), and Cu (st. 112, 240).

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Ekaterina A. Kotelyanets – data preparation, analysis and synthesis of research results, presentation of data in the text and their analysis, article correction, advisory assistance

Yulia S. Gurova – data processing, analysis and interpretation of data, participation in the discussion of the paper materials, paper correction, formulation of the conclusions

Oleg V. Stepanyan – participation in the discussion of the paper materials, paper correction

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