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Assessment of the Ability of Suspended Matter in the Sea of Azov to Concentrate Heavy Metals

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Abstract
With a large specific surface area, suspended matter can concentrate heavy metals to high levels. Falling down by gravity, suspended particles can deposit pollutants into the bottom sediments thus participating in the self-purification of sea water. The purpose of this work is to assess the ability of suspended matter in the Sea of Azov to concentrate Pb, Zn, Cu, Cd, and Hg. For the purpose of the study, two areas of the Sea of Azov were identified (Taganrog Bay and the central part of the sea) given their morphometric and hydrological features. Mass concentrations of Pb, Zn, Cu, Cd were determined by the electrothermal atomic absorption method; measurements of the mass concentration of Hg were carried out by the method of flameless atomic absorption spectrometry. The content of each metal in suspended matter was calculated in relation to that in water based on the accumulation factors. The obtained accumulation factors indicate a high ability of suspended matter to concentrate mercury, copper and zinc. The concentration of cadmium in the suspended matter was insignificant. This is because in surface waters cadmium migrates mainly in a dissolved state, with the suspended forms normally not exceeding 20–30 %. The content of lead in the suspended matter did not exceed 12.4 % in the central part of the sea and 15.8 % in Taganrog Bay, both of its total content. It is shown that when the values of the factors of heavy metal accumulation by suspended matter exceed 105, almost the entire volume of the studied heavy metals is in the suspended matter. These data indicate the high significance of the suspended matter concentrating ability for self-purification of water from heavy metals

Keywords: Sea of Azov, suspended matter, mercury, lead, cadmium, copper, zinc, accumulation factor

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Оценка способности взвесей Азовского моря концентрировать тяжелые металлы

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Аннотация

Обладая большой удельной поверхностью, взвешенное вещество может концентрировать тяжелые металлы до высоких уровней. Опускаясь в результате гравитации, взвешенные частицы могут депонировать загрязнения в толщу донных отложений, таким образом участвуя в самоочищении морской воды. Цель работы заключалась в оценке способности взвесей Азовского моря концентрировать Pb, Zn, Cu, Cd и Hg. Для исследования были выделены два района Азовского моря – Таганрогский залив и центральная часть моря, что связано с их морфометрическими и гидрологическими особенностями. Массовые концентрации Pb, Zn, Cu, Cd определялись электротермическим атомно-абсорбционным методом; измерения массовой концентрации Hg проводились методом беспламенной атомно-абсорбционной спектрометрии. Содержание каждого металла во взвеси по отношению к его содержанию в воде было рассчитано на основе коэффициентов накопления. Полученные коэффициенты накопления свидетельствуют о высокой способности взвесей концентрировать ртуть, медь и цинк. Концентрирование кадмия на взвесях было незначительным, это связано с тем, что в поверхностных водах кадмий мигрирует в основном в растворенном состоянии, взвешенные формы, как правило, не превышают 20–30 % от его валового содержания. Содержание свинца во взвешенном веществе не превышало 12.4 % в центральной части моря и 15.8 % в Таганрогском заливе от общего его содержания. Показано, что при значениях коэффициентов накопления тяжелых металлов , бóль 10^5, практически весь объем исследуемых тяжелых металлов находится на взвеси. Эти данные свидетельствуют о высокой значимости фактора концентрирующей способности взвесей в самоочищении вод от тяжелых металлов.

Ключевые слова: Азовское море, взвешенное вещество, ртуть, свинец, кадмий, медь, цинк, коэффициент накопления

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Introduction

A characteristic feature of the biospheric cycles of mass exchange is their discontinuity, which manifests itself in the imbalance of masses at the so-called input and output of the cycle. The removal of an excess part of the migrating masses from the migration cycle with their accumulation in any component of the natural environment (in sediments, soil, a large body of water, etc.) or with their redeployment to another migration flow maintains the steady state of open non-equilibrium natural systems and the direction of their development [1)].

When studying the biogeochemical features of the behavior of heavy metals in marine ecosystems, two approaches are employed. The first one is based on the determination of the concentration of metals in various components of ecosystems. The second one involves the assessment of metal flows caused by the physical movement of matter and biogeochemical processes that change this matter. In real practice, these are interrelated characteristics, since the system concentration changes only when the incoming flow is not equal to the outgoing one. Any change in the concentration of the metal results in the occurrence of a corresponding flow. It should be noted that the converse is not always true, i.e., it is quite common when very intense flows of metals in the system are not accompanied by a change in concentration. This occurs when the incoming flow is equal to the outgoing one. Ideally, to study the behavior of metals in marine ecosystems, it is necessary to create and use models that combine concentration and flow characteristics [2)].

Unlike most organic pollutants subject to gradual destruction, heavy metals are just redistributed among various components of ecosystems. The main biogeochemical mechanism of self-purification of sea water from heavy metals is its sedimentation by suspended matter [1, 2]. With a large specific surface area, suspended matter can concentrate heavy metals to high levels. Moving downwards due to gravity, suspended particles can deposit pollutants into the bottom sediments, thus participating in the self-purification of sea water [1)].

Dissolved forms of metals entering the coastal area with river runoff, slope wash or aerially, are assimilated by phytoplankton and sorbed on various suspended particles. During the destruction of plankton, some elements go back into solution. The metals remaining in the solid either settle down or pass through the food chain into zooplankton. The metals absorbed by zooplankton also partially go back into solution and partially settle down as a part of organomineral aggregates. At the very bottom and in the upper layer of bottom sediments, a complex of biogeochemical processes takes place with both the mobilization of some metals into pore and bottom water and the reverse binding of metals into sulfides and hydroxides formed. The transformation and destruction of organic matter represent the engine of growth for the transformation of metals at the bottom and in sediments.

[1)] Dobrovolsky, V.V., 1999. Range of Masses of Dispersed Chemical Elements Migrating in the Soil-Vegetation System in Zonal Phytocenoses of the World Land. In: GEOKHI RAS, 1999. *Proceedings of the Second Russian School Geochemical Ecology and Biogeochemical Zoning of the Biosphere, Moscow, 25-28 January 1999*. Moscow: GEOKHI RAS, pp. 32–33 (in Russian). Polikarpov, G.G. and Egorov, V.N., 1986. [*Marine Dynamic Radiochemoecology*]. Moscow: Energoatomizdat, 176 p. (in Russian).

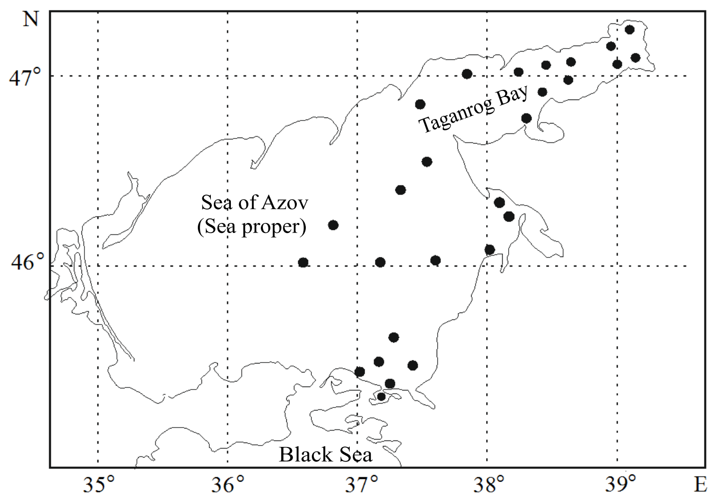
[2)] Shul'kin, V.M., 2007. [*Heavy Metals in River and Coastal Marine Ecosystems. Dissertation of the Doctor of Geographical Sciences*]. Vladivostok: Pacific Institute of Geography DO RAS, 289 p. (in Russian).

As judged by rather low content of most dissolved metals in sea and, moreover, in ocean water compared to river water, the general trend in the behavior of most heavy metals represents their removal and binding as a part of the settling material and further in bottom sediments. Metals, initially entering the sea with terrigenous suspended solid material, mainly continue their migration as its part, although at high concentration of mobile forms of metals in terrigenous suspended solid material, their desorption into water is possible with increased salinity in estuaries [3].

A great number of works have been devoted to the study of the content of heavy metals in water and bottom sediments of the Sea of Azov in different years [2, 4–8]. The results of the study by A.V. Mikhailenko (2018) on establishing patterns of spatial distribution of heavy metal concentrations in water and in the suspended matter of the Sea of Azov, are advantageous [5]. Since suspended matter can concentrate heavy metals to high levels due to its large specific surface, the assessment of the accumulation of Pb, Zn, Cu, Cd and Hg in suspended solid material is considered to be a relevant study. The aim of this study is to assess the ability of suspended solid material to concentrate mercury, copper, cadmium, zinc and lead in the spring-and-summer and autumn-and-winter periods in the central part of the Sea of Azov and in Taganrog Bay. It continues the series of studies established by [9].

Materials and methods

The work used data on the concentration of Pb, Zn, Cu, Cd, Hg in the water and in the suspended matter of the Sea of Azov in 2015, provided by *Azovmorinformtsentr* Federal State Budgetary Institution in cooperation with the Department of Ecology and Nature Management of MGRI. For analysis, water samples were taken using *PE-1220* sampling system according to GOST R 51592-2000 in the surface (0–5 m) layer at 27 points (Fig.) in spring (March and April), summer (June and July), autumn (September and October) and winter (December). Unfiltered samples (total form) were used to determine metals.



Sampling points

Chemical analysis of water samples concerning lead content was carried out in accordance with the method stipulated by Federal Environmental Regulatory Documents PND F 14.1:2:4.140-98, the limit of detection made 0.0002 mg/L; cadmium – PND F 14.1:2:4.140-98, the limit of detection – 0.00001 mg/L; copper – PND F 14.1:2:4.140-98, the limit of detection – 0.0001 mg/L; zinc – M-MVI-539-03, the limit of detection – 0.001 mg/L; mercury – PND F 14.1:2:4.260-2010, the limit of detection – 0.01 mg/L. The concentrations of the above stated heavy metals were measured using *KVANT-Z-ETA* atomic absorption spectrometer.

In the work, two areas were identified in the Sea of Azov – Taganrog Bay and the open water area of the Sea of Azov (the sea proper), which was associated with their morphometric and hydrological features.

Table 1 shows the threshold limit values of the heavy metals under consideration in the sea water.

Literature data [5] were also used to determine the concentration of heavy metals in the suspended matter of the Sea of Azov.

The accumulation factors of heavy metals by suspended matter (K_{sm}) were calculated using the following equation

$$K_{sm} = \frac{1000 \cdot C_{sm}}{C_w},$$

where C_{sm} – concentration of heavy metal in suspended matter, μg/g, based on dry weight; C_w – concentration of heavy metal in water, μg/L [1].

The accumulation factors were calculated based on the results of observations with an accuracy of three significant figures, which corresponded to an error with the value not more than 0.1 %.

The content of heavy metal in suspended solid material (P_{ssm}) in relation to its content in the marine environment was calculated by the following formula [1]

$$P_{ssm} = \frac{m_{sp} \cdot K_{sm} \cdot 100}{m_{sp} \cdot K_{sm} + 1} \ (\%), \qquad (1)$$

where m_{sp} – specific mass of suspended solid material in water, g/m^3.

Table 1. Threshold limit values of heavy metals in water (TLV_w) [3)]

Parameter	Metal				
	Pb	Zn	Cu	Cd	Hg
Class of hazard	3	3	3	2	1
TLV_w, μg/L	10.0	50.0	5.0	10.0	0.1

[3)] On the Approval of Water Quality Standards for Water Bodies of Commercial Fishing Importance, Including Standards for Maximum Permissible Concentrations of Harmful Substances in the Waters of Water Bodies of Commercial Fishing Importance: Order of the Ministry of Agriculture of Russia dated December 13, 2016, No. 552. URL: http://publication.pravo.gov.ru/Document/View/0001201701160006 (Accessed: 25.02.2022).

Results and discussion

Table 2 shows that in the observed period of time (2015), the concentrations of Hg, Pb, Zn and Cu in many samples exceeded the TLV. Attention is drawn to the average values of copper concentration in water, which exceeded the TLV in all seasons in the sea proper and in Taganrog Bay, as well as high concentrations of zinc in Taganrog Bay. High concentrations of copper were also observed in the suspended matter (Table 3).

Suspended matter is a complex polydisperse multicomponent system. The material composition of the suspended solid material in the Sea of Azov is characterized by considerable diversity. According to the genetic trait, mineral components of terrigenous and chemogenic origin, as well as organic residues at various stages of mineralization, are distinguished in the suspended material. As it is shown by Yu.P. Khrustalev (1981) studies, during the year the terrigenous component of suspended solid material prevails [4)]. The terrigenous type of suspended matter is characterized by the predominance of products of coastal abrasion and river runoff (more than 70 %) in its composition

T a b l e 2 . Content of heavy metals in water of the Sea of Azov in 2015 (total form), μg/L

Area	Hg	Cu	Cd	Zn	Pb
spring and summer					
Sea proper (15)	0.003–0.283 0.045	4.0–23.8 10.10	0.1–1.4 0.41	7.3–34.0 31.60	3.9–34.0 12.18
Taganrog Bay (12)	0.002–0.042 0.034	9.2–20.5 14.1	0.2–1.7 0.89	8.0–127.0 57.2	1.0–13.0 4.46
autumn and winter					
Sea proper (15)	0.004–0.460 0.069	27.0–27.3 8.7	0.2–1.7 0.82	7.1–74.1 35.1	5.4–11.0 7.9
Taganrog Bay (12)	0.008–0.130 0.061	1.6–27.7 8.8	0.01–6.6 1.44	9.7–120.0 34.2	3.4–17.0 7.45

Note: in brackets – number of samples; above the line – concentration range; under the line – average value.

4) Khrustalev, Yu.P., Ganicheva, L.Z. and Volkova, E.N., 1981. Geochemistry of Suspended Sediment of the Sea of Azov. In: Yu. P. Khrustalev, 1981. *Geographical Aspects of Hydrological and Hydrochemical Study of the Azov Basin*. Leningrad: GO USSR, pp. 76–87 (in Russian).

Table 3. Content of heavy metals in the suspended matter of the Sea of Azov in 2015, μg/L [5, p. 103]

Area	Hg	Cu	Cd	Zn	Pb
spring and summer					
Sea proper	0.001–0.234 0.043	1.0–19.0 9.6	0.03–0.09 0.035	3.6–50.7 29.2	2.94–3.10 1.1
Taganrog Bay	0.005–0.027 0.013	0.3–31.1 11.2	0.01–0.14 0.06	4.3–83.4 31.5	0.72 –1.58 1.32
autumn and winter					
Sea proper	0.001–0.09 0.020	0.3–31.1 7.5	0.02–0.6 0.14	3.1–80.1 31.6	0.21–2.77 0.98
Taganrog Bay	0.003–0.314 0.055	0.4–29.3 7.7	0.03– 0.7 0.05	3.3–79.4 27.0	0.27–1.48 1.18

Note: above the line – concentration range; under the line – average value.

and is represented mainly by pelitic fractions. The content of aleuritic particles increases (up to 50 %) in the delta-front areas of the rivers during the flood period and during intense waves in the coastal area [10, p. 52].

In the Sea of Azov, there is an area of constantly high content of suspended matter (Taganrog Bay) and an area of its lower concentrations (the sea proper). It should be noted that under conditions of wind activity, the content of suspended matter can increase by a factor of 2–6. In this case, such changes in concentration depend on the nature of the soil in the area of waves. Bottom sediments of muddy bottom are mobilized most intensively into the water layer [11].

In the vertical distribution of suspended particles, a clear increase in their number from the surface horizon of the water layer to the bottom one was established. According to the averaged data, the concentration of suspended solid material in the bottom layer makes 111.0–165.8 mg/L. The amount of suspended solid material in the bottom layer is determined by the level of sedimentation processes, the nature of the soil and its predisposition to the spreading of sediments. The maximum values of suspended solid material concentrations are noted in the bottom horizon above clayey silts (218–229 mg/L), and the minimum values are noted above sandy and shelly soils (83.5–87.4 mg/L)[5)].

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5) Mirzoyan, Z.A., 1984. [*Suspended Sediment in the Sea of Azov and Its Role in Feeding Plankton and Benthic Animals. Dissertation of the Candidate of Biological Sciences*]. Rostov-on-Don : AzNIIRKH, 168 p. (in Russian).

T a b l e 4 . Factors of heavy metal accumulation by suspended matter (K_{sm}) and content of heavy metals in suspensions (P_{ssm}), %, in relation to their content in the aquatic environment

Area	Hg	Cu	Cd	Zn	Pb
spring and summer					
Sea proper	1.132 / 95.6	1.011 / 95.0	0.005 / 8.3	0.640 / 92.4	0.005 / 9.1
Taganrog Bay	0.016 / 38.2	0.099 / 79.4	0.002 / 6.7	0.031 / 55.1	0.011 / 29.6
autumn and winter					
Sea proper	0.021 / 29.0	0.329 / 86.2	0.011 / 17.1	0.475 / 90.0	0.007 / 12.4
Taganrog Bay	0.235 / 90.2	0.179 / 87.5	0.001 / 3.5	0.096 / 78.9	0.005 / 15.8

Note: above the line – $K_{sm} \cdot 10^6$; under the line – P_{ssm}

Table 4 shows the results of the calculation of the accumulation factor of heavy metals by suspended solid material and the content of heavy metal in suspensions in relation to its content in the marine environment.

The obtained accumulation coefficients indicate a high ability of suspensions to concentrate mercury, copper and zinc in the spring-and-summer and autumn-and-winter periods both in Taganrog Bay and in the sea proper. The calculated content of heavy metals in suspended solid material ranged from 29 to 95.6 % of their total content in the marine environment.

Similar results are presented in the studies by A.P. Stetsyuk and V.N. Egorov (2018), in which the dependencies of mercury concentration by suspended matter of the Crimean shelf of the Black Sea were determined. In [12, p. 3], the coefficients of the ability of suspensions to concentrate mercury were in the range of $0.023 \cdot 10^6$–$7.067 \cdot 10^6$, and the mercury content on suspensions reached 98 % of the total mercury content in the marine environment.

The content of cadmium in suspended solid material was insignificant and made 3.5–6.7 % for Taganrog Bay and 8.3–17.1 % for the sea proper. This may be resulted from the fact that cadmium migrates in surface water mainly in the dissolved state; suspended forms, as a rule, do not exceed 20–30 % [6)].

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6) Perelman, A.I. and Kasimov, N.S., 2000. *Landscape Geochemistry*. Moscow: MGU, 565 p. (in Russian).

Lead is characterized by a low degree of solubility, which determines its entry into the river runoff of the Don and the Kuban mainly in suspended state. The accumulation factors of the lead by suspended solid material, and, consequently, its content in suspended solid material, were expected to be higher. Nevertheless, our results show that the lead content in suspended matter does not exceed 12.4 % in the sea proper and 15.8 % in Taganrog Bay of its total content in the marine environment. This situation can be explained by the fact that the value of the accumulation factor by suspended solid material depends on the concentration of heavy metal in water, as at low values of C_w, the accumulation factor increases, and with an increase in C_w, it decreases. The concentration of lead in the central part of the sea and in Taganrog Bay during the study period was high, often exceeding the TLV by 2–3 times. This fact can confirm the effect of saturation of suspended solid material with lead. Similar dependences were obtained in the study of the sorption capacity of the bottom sediments of the Sea of Azov concerning lead. The materials shown in the work of Academician G.G. Matishov (2017) demonstrated that the increased intensity of sedimentary self-purification of water at low concentration of lead in water was provided by high (at $K_l > n \cdot 10^4$ units) concentrating ability of bottom sediments. With an increase in the degree of water pollution with lead to MPC (10 μg/L), the K_l value decreased by more than two orders of value and, accordingly, the contribution of sedimentation processes to water self-purification decreased [2].

In general, the results presented in this work show that the accumulation factor can be interpreted as an important indicator of the intensity of biogeochemical cycles of marine environment pollutants.

Conclusions

The obtained accumulation factors indicate a high ability of suspensions to concentrate mercury, copper and zinc in the spring-and-summer and autumn-and-winter periods both in Taganrog Bay and in the sea proper. The conducted studies show that with the values of the accumulation factors $K_{sm} > 10^5$, almost all the content of the studied heavy metals is located in suspended solid material. These data confirm the high importance of the factor of the concentrating ability of suspensions in the self-purification of water from heavy metals.

The content of cadmium in suspended solid material was insignificant, up to 6.7 % in the Taganrog Bay and up to 17.1 % in the sea proper, which can be explained by the weak complexing ability of cadmium compared to other metals.

High concentrations of lead in the water of the Sea of Azov can determine the low accumulation factors of this metal in suspended solid material. This can confirm the fact of saturation of suspended solid material with lead. Thus, the significance of the concentrating ability of suspended solid material depends on the concentration of heavy metals in water. At low values, it prevails over other biogeochemical mechanisms of water self-purification and decreases with increasing concentration.

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The author has read and approved the final manuscript.