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MERCURY ACCUMULATION IN SUSPENDED MATTER OF FOAM AND WATER OF THE BLACK SEA*

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The ability of suspended matter to concentrate mercury may be the prevailing factor in Black Sea purification. As a result of sedimentation, suspended particles transport pollution from the surface layer of the water column and, as a consequence, can deposit them in bottom sediments, thus participating in self-purification of marine area. Suspended matter, as a dispersed phase of an aqueous medium, considered as a heterogeneous dispersed system, can be more saturated with mercury than water itself, as a dispersion medium. In this work, contribution of dissolved and suspended forms of mercury to its total content was determined, and concentrating ability of suspended matter relative to mercury, which affects biogeochemical self-purification of waters from mercury, was estimated. All water samples were separated into filtrate and suspension by filtration through nucleopore filters with a pore diameter of 0.45 μm. Measurements of mercury concentration were carried out using a Hiranuma-1 analyzer by the method of atomic absorption spectrophotometry. Concentration of dissolved mercury in water was determined per liter, while in suspended matter – per liter and per gram of dry weight. Prevalence of dissolved form of mercury was revealed regardless of the season, with its percentage varying from 66.3 to 85.8 % of total mercury concentration. Average content of suspended form varied in the range of 14.2–33.7 % of its total form. Values of the dry weight of suspended matter (m_{ss}) varied from 0.1 to 15.0 mg·L^{-1} over the entire period studied, and an accumulation coefficient of mercury in suspended matter (K_{ss}) varied from n·10^3 to n·10^7. Significant contribution of suspended form of mercury in sea foam to its total content in stormy weather was established. With dry weight of suspended matter in seawater reaching 9.6 mg·L^{-1}, the concentration of dissolved form of mercury reached 55 ng·L^{-1}, and the concentration of suspended one reached 20 ng·L^{-1}. In sea foam, the concentration of suspended sedimentary matter was of 895.2 mg·L^{-1}; mercury concentration reached 200 ng·L^{-1} in dissolved form and 260 ng·L^{-1} in suspended one. Total mercury concentration in sea foam in this case exceeded the threshold limit value (100 ng·L^{-1}) for seawater. The accumulation coefficient of mercury in suspended matter (K_{ss}) was 3.8·10^4 for seawater and 1.5·10^3 for foam. Such distribution of mercury in sea suspension, foam, and water, as well as K_{ss} values obtained, may indicate high significance of suspended matter in self-purification of marine area. At a low mercury content in water, the concentrating ability of suspended matter, characterized by relatively high values of its mercury accumulation coefficient, becomes a very significant factor in the sedimentation self-purification of waters from mercury; however, with an increase in water pollution with mercury, the effect of this factor decreases.

Keywords: mercury, suspended matter, sea foam, Black Sea

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Mercury is a substance of the I hazard class and one of the most environmentally significant pollutants of the Black Sea [14]. As known, at mercury concentration of 0.1 μg·L^{-1}, vital activity of unicellular algae is suppressed [12]. At mercury concentration of 0.018 mg·L^{-1}, biochemical processes of self-purification are inhibited in waters, and at 2.0 mg·L^{-1} they stop [1 ; 4].

Mercury can enter the environment from various natural and anthropogenic sources. Volcanic activity, degassing of the Earth's crust, and evaporation from water surface followed by wind transport can be considered the main mercury natural sources. With Danube River waters only, mercury inflow into the Black Sea is estimated at 48.7–58.9 t·year^{-1} [16], and its inflow into entire marine area from other sources – at 80 t·year^{-1} [17]. Mercury anthropogenic sources in the Black Sea include industrial and municipal wastewater, ship repair facilities, and marine transport.

According to data published, the predominant factor in sedimentary water purification is a concentrating ability of suspensions [2 ; 10]. Due to sorption, mercury concentration in suspended matter can be quite considerable. As a result of sedimentation, suspended particles transport pollution from the surface layer of the water column and, as a consequence, can deposit them in bottom sediments, thus participating in self-purification of marine area. In stormy weather (mainly in shallow shelf and coastal areas), the upper layer of bottom sediments mixes with the bottom water layer; due to biogeochemical cycle, pollution inflows back into the water column. At the same time, surface water of a coastal water area is intensively filled with suspended sediment and is therefore a qualitatively new convenient object for studying the processes of self-purification of marine environment from mercury. Dispersed sedimentary matter, like almost all biotic and abiotic components of marine ecosystems, can be more saturated with mercury compared to water, considered as a dispersion medium.

The aim of this work was to determine the contribution of dissolved and suspended forms of mercury to its total content and the concentrating ability of suspended matter relative to mercury, as well as to estimate percentage of mercury accumulation by suspension, which characterizes contribution of suspended form of mercury to total form in a "suspension – water" system. A data array on mercury content in the Black Sea in 2011–2017 was analyzed, and mercury concentration in sea foam in 2018 was studied.

MATERIAL AND METHODS

To identify the contribution of dissolved and suspended forms of mercury to total one, published and unpublished data on mercury content in the Black Sea for 2011–2017 were used [9 ; 10]. Sampling stations are shown in Fig. 1. Water for the study was sampled during research cruises of the RV "Professor Vodyanitsky" (No. 70, 72, 79, 80, 88, 90, 92, 93, 96, 99, and 100), as well as in Sevastopol bays. In stormy weather in 2018, water and foam samples were taken from the surface layer of the Black Sea (44°39.167′N, 31°50.445′E).

All water samples were separated by filtration through pre-weighed nucleopore filters with a pore diameter of 0.45 μm into two parts: filtrate and suspension on the filters. Then, the suspension was dried under natural conditions and again weighed using a Sartorius microanalytical balance with a measurement error of 0.1 mg. Dry weight of the suspended matter was calculated from the difference between the filter mass after and before filtration. After that, the filter with the dry suspension was processed according to GOST 26927 (All-Union State Standard). Measurements of mercury concentration were carried out by flameless atomic absorption spectrophotometry using a Hiranuma-1 analyzer. To calibrate it, certified standard samples of mercury ion solution GSO 7879-2001 were used. Analysis of several calibration solutions with mercury concentration of 0.2, 0.4, 0.6, 0.8, and 1 $\mu g \cdot L^{-1}$ (10 replicates each) showed a reproducibility of the results with a relative error not exceeding 2 %.

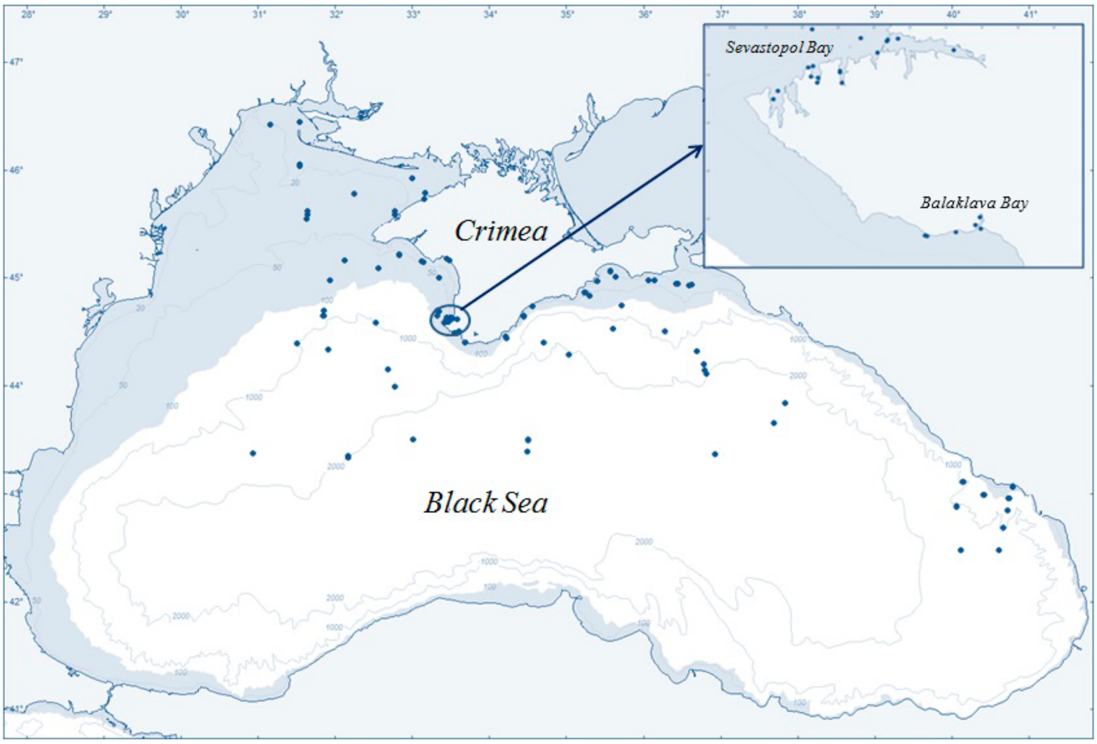

Fig. 1. Map of sampling in the Black Sea [13]

Concentration of dissolved form of mercury in water was determined per liter, and in suspended matter – per liter and per gram of dry weight.

To characterize the concentrating ability of suspended matter, the accumulation coefficient of mercury (K_{ss}) was calculated [7]:

$$K_{ss} = \frac{1000 \cdot C_{ss}}{C_w} , \qquad (1)$$

where C_{ss} is specific mercury concentration in suspended matter (suspended substance), $ng \cdot g^{-1}$;

C_w is concentration of dissolved form of mercury in water, $ng \cdot L^{-1}$.

Dependence of the percentage of mercury accumulation by suspended matter from aquatic environment on accumulation coefficients (K_{ss}) and concentration of suspended matter $10^{-6}B$ was calculated by the formula [6]:

$$A_1 = \frac{K_{ss}}{K_{ss} + \frac{1}{10^{-6}B}} (\%) , \qquad (2)$$

where $B = \frac{P_1}{P_2}$;

P_1 is mass of dry suspension;

P_2 is mass of water equal to 10^6 g;

$10^{-6}B$ is suspension concentration in the aquatic environment in parts per million ($mg \cdot L^{-1}$).

RESULTS AND DISCUSSION

Data for 2011–2017 were analyzed by season and depth. For each data group, average concentrations of various forms of mercury in ng·L^{-1} were calculated (Fig. 2). The maximum average annual concentration of total form of mercury was registered in 2014, and the minimum one – in spring of 2012 (Fig. 2A). The results showed the prevalence of dissolved form of mercury (Fig. 2B) with a variation in its percentage in the range of 66.3–85.8 % of total mercury concentration. The average concentration of suspended form was 14.2–33.7 % of the concentration of its total form.

In this case, the values of the dry weight concentration of suspended matter varied 0.1–15.0 mg·L^{-1} over the entire period studied.

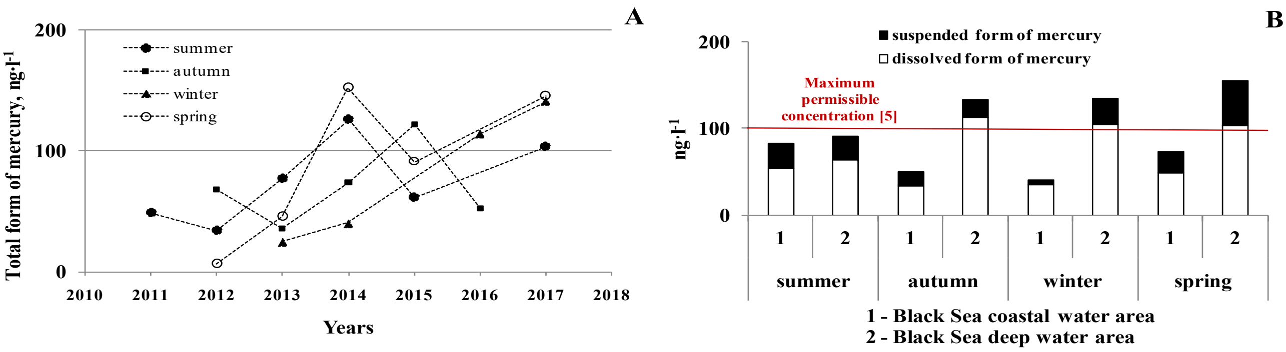

Fig. 2. A – average annual concentration of total form of mercury in the Black Sea in 2011–2017; B – average concentrations of dissolved and suspended forms of mercury during different seasons

Fig. 2B shows that in summer, both in coastal and deep-water areas of the Black Sea, the average concentrations of dissolved and suspended forms of mercury are approximately equal, with total content not exceeding threshold limit value (hereinafter TLV) for seawater equal to 100 ng·L^{-1}. In other seasons of the year, for coastal water areas of the Black Sea, the average mercury concentration is comparatively lower, and for deep-water areas it is much higher. Fig. 2B shows that in spring, the average concentrations of dissolved and suspended forms of mercury, both in coastal and deep-water areas, are higher compared to those in autumn and winter. An increased contribution of suspended form of mercury to its total content is also observed, which indicates activation of accumulating ability of the suspension relative to mercury. This is most likely to be due to spring increase of primary producing ability of biotic component of the suspension, in particular phytoplankton, resulting from increasing impact of two factors: heat and light. The increased concentration of dissolved mercury in deep-water areas of the Black Sea during these three relatively low-temperature seasons (autumn, winter, and spring), compared to coastal waters, is most likely associated with the low accumulating ability of suspended matter relative to mercury and with the corresponding reduced eliminating ability of the suspension to transport mercury by sedimentation from the surface layer to the bottom since it was previously described in literature that total suspension of surface water in the Black Sea and the Sea of Azov near the Crimean Peninsula is regularly decreasing from coastal to open deep-sea waters [8].

The prevalence of the values of suspended and dissolved forms of mercury in summer season, both for coastal and deep-water areas of the Black Sea, over mercury content in coastal waters in relatively low temperature seasons (autumn, winter, and spring) is most likely associated with an increase in the primary producing ability of biotic component of the suspension (mainly phytoplankton) [3] and with a corresponding increase in its eliminating ability to transport accumulated mercury from the surface layer towards bottom sediments.

Table 1 shows the average values for the entire data array, as well as the ranges of variation of suspended matter specific dry mass in different seasons of the year. The minimum content of suspended matter was registered in summer (0.99 mg·L^{-1}) and winter (0.60 mg·L^{-1}) seasons in deep-water areas of the Black Sea. The maximum average values of suspended matter (m_{ss}) were observed in winter (2.96 mg·L^{-1}) and spring (3.90 mg·L^{-1}) periods in Crimean coastal waters.

Table 1. Average values of suspension concentration and specific mercury concentration in suspension from Black Sea water area in 2011–2017 (figures in brackets indicate ranges of variation)

Season of the year	Area under study	Suspension concentration (m_{ss}), mg·L^{-1}	Specific mercury concentration in suspension (C_{ss}), ng·g^{-1} of dry weight
Summer	Coastal water area of Crimea	1.75 (0.3–10)	30,197 (408–500,000)
	Deep-water area of the Black Sea	0.99 (0.1–3.1)	53,038 (4839–320,000)
Autumn	Coastal water area of Crimea	1.67 (0.6–7)	9625 (614–30,769)
	Deep-water area of the Black Sea	1.08 (0.2–3)	23,353 (1447–100,000)
Winter	Coastal water area of Crimea	2.96 (0.5–14.7)	5144 (314–41,667)
	Deep-water area of the Black Sea	0.60 (0.1–3.4)	131,317 (3333–1,100,000)
Spring	Coastal water area of Crimea	3.90 (0.5–15)	17,375 (825–85,000)
	Deep-water area of the Black Sea	1.62 (0.5–3.6)	10,831 (2861–26,400)

Based on suspension weight (g·L^{-1}) and mercury concentration in it (ng·L^{-1}), mercury concentration in suspended matter (ng·g^{-1}) was calculated (see Table 1). The minimum mercury concentration in suspension was 314 ng·g^{-1} in coastal water area of Crimea in winter. The highest value (1,100,000 ng·g^{-1}) was registered in deep-water area of the Black Sea in winter. Calculated by formula (1), the accumulation coefficient of mercury in suspended matter (K_{ss}) varied from $n \cdot 10^3$ to $n \cdot 10^7$.

Fig. 3 shows a graphical depiction of a dependence of change in the accumulation coefficient of mercury in suspended matter (K_{ss}) on its concentration in water (C_w) for different seasons of the year and water areas. The graph (Fig. 3D) shows that in spring in coastal area of the Black Sea, the values of K_{ss} varied with an increase in C_w with a statistical validity of $R^2 = 0.73$. In the deep-water areas of the Black Sea in the same season, the dependences of K_{ss} on C_w had a low coefficient of approximation validity ($R^2 = 0.04$). In summer, R^2 values amounted to 0.01 for coastal water area of the Black Sea and 0.32 for deep-water area (Fig. 3A). In autumn, this parameter had the value 0.64 for coastal water area and 0.25 for the deep-water one (Fig. 3B). In winter, $R^2 = 0.01$ for deep-water areas of the Black Sea and $R^2 = 0.50$ for coastal water area (Fig. 3C). As a result, the most reliable trends were identified for coastal waters, with the exception of summer season.

The dependences obtained (Fig. 3) had a low coefficient of determination in summer in coastal water area (Fig. 3A); it was also low in winter (Fig. 3C) and spring (Fig. 3D) in the deep-water area. This indicates different representativeness of the data, as well as different ability of suspensions to accumulate mercury from coastal and deep-water areas in different seasons of the year. In general, by the approximating dependence in other cases, we can say that with an increase in the concentration of dissolved form of mercury in water (C_w), concentrating ability of suspended matter decreases.

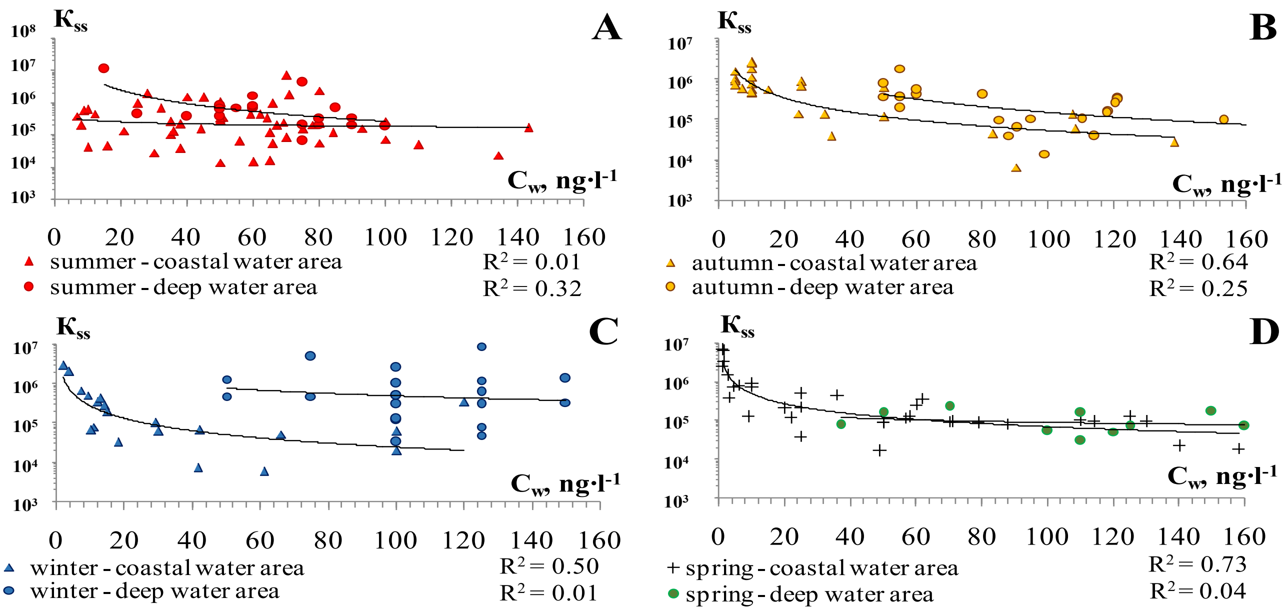

Fig. 3. Dependence of change of the accumulation coefficient of mercury in suspended matter (K_{ss}) on concentration of dissolved form of mercury in water (C_w, ng·L^{-1}) during different seasons (A – summer; B – autumn; C – winter; and D – spring) and in different water areas

Using formula (2), a dependence of the percentage of mercury accumulation by suspended matter from marine environment on the accumulation coefficient was calculated (Fig. 4).

According to Fig. 4, the percentage of mercury accumulation by suspension varies with an increase in K_{ss} with almost the same statistical validity for different seasons and Black Sea areas, with the exception of spring season in deep-water area. According to the data obtained, with $K_{ss} > 10^6$, almost all mercury is accumulated by suspended matter of seawater, which indicates its high concentrating ability.

The dependence of the percentage of mercury accumulation by suspension on C_w for different seasons and water areas has slightly pronounced trends (Fig. 5), with the exception of spring period in the deep-water area ($R^2 = 0.83$) (Fig. 5D). Moreover, in all cases, there is a decrease in a ratio of suspended form of mercury with an increase in the concentration of dissolved form in water, which is in full agreement with the functional dependence expressed by formula (2).

Previously, when studying the mechanism of foam formation on seawater surface, it was found that organic matter of seawater transforms from dissolved state into foam and suspension since fragments of the shells of bursting foam bubbles turn into dispersed particles [11].

In this article, the effect of storm surge on ratio and magnitude of different forms of mercury in water was investigated. In stormy weather, the concentration of suspended sedimentary matter in seawater was 9.6 mg·L^{-1}, and in sea foam – 895.2 mg·L^{-1} (Fig. 6).

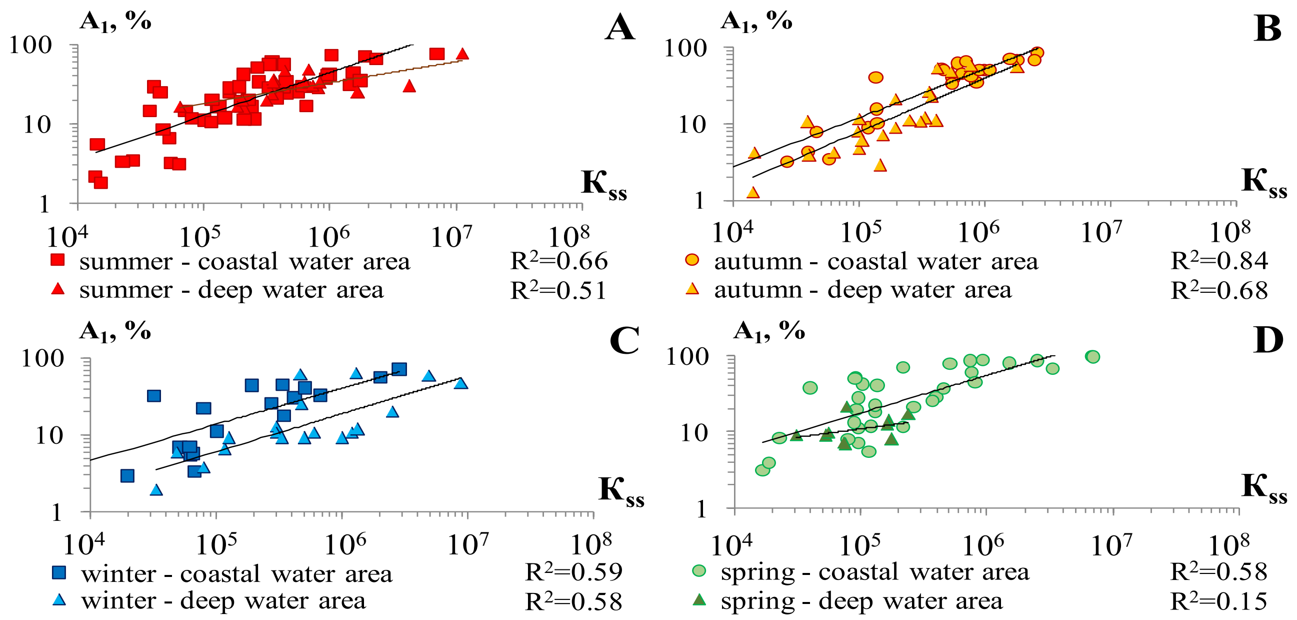

Fig. 4. Dependence of the percentage of mercury accumulation by suspended matter (A_1, %) on the accumulation coefficient (K_{ss}) during different seasons (A – summer; B – autumn; C – winter; and D – spring) and in different water areas

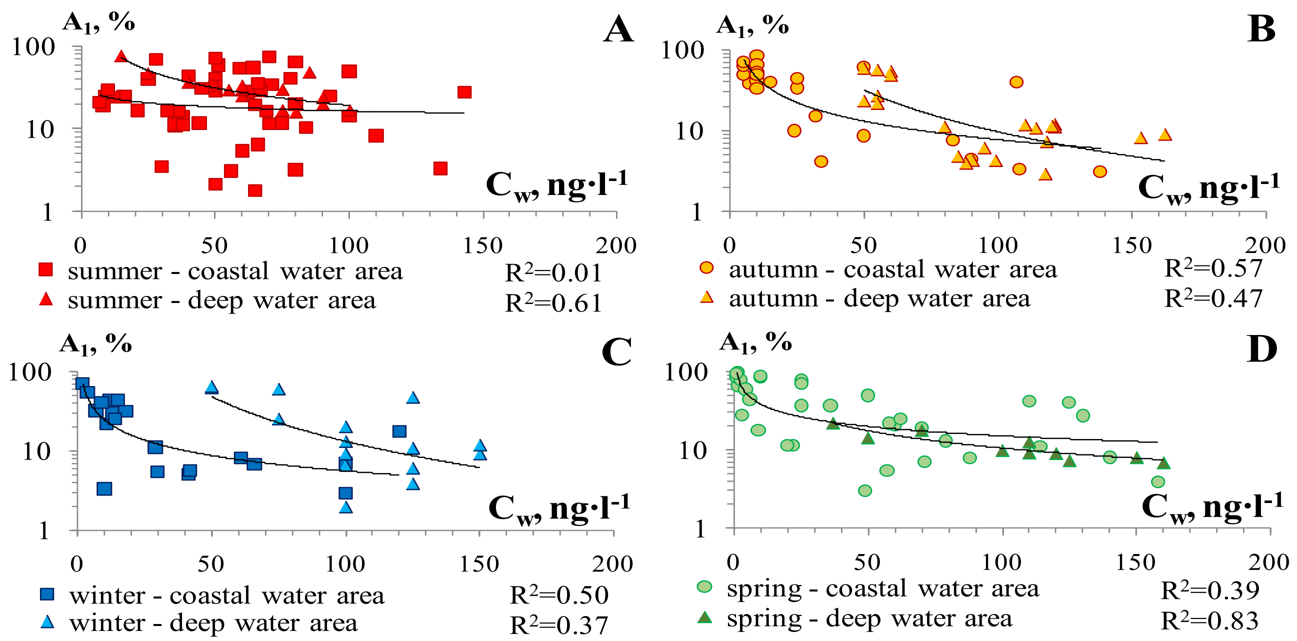

Fig. 5. Dependence of the percentage of mercury accumulation by suspended matter (A_1, %) on dissolved form of mercury (C_w, ng·L^{-1}) during different seasons (A – summer; B – autumn; C – winter; and D – spring) and in different water areas

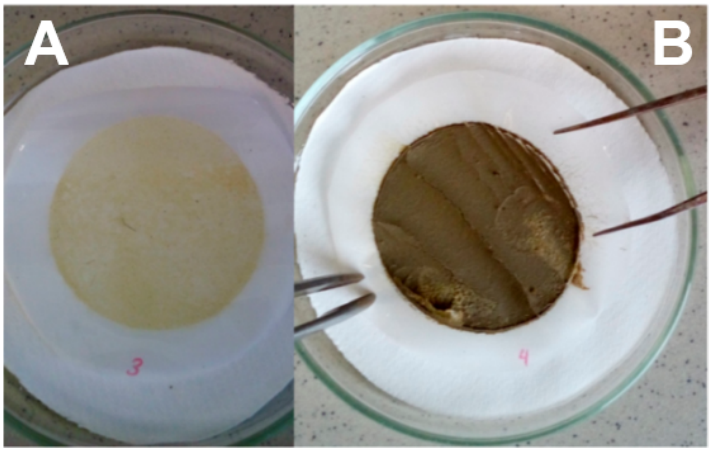

Fig. 6. A – suspended matter obtained from seawater; B – suspended matter obtained from sea foam

The concentration of dissolved form of mercury in seawater was 55 ng·L^{-1}, and in sea foam it reached 200 ng·L^{-1}, exceeding TLV (100 ng·L^{-1}) [5]. Values of suspended form of mercury were 20 ng·L^{-1} in seawater and 260 ng·L^{-1} in sea foam (Fig. 7). In this case, the concentration of total mercury in sea foam exceeded not only TLV for seawater but also standards for bottom sediments (300 ng·L^{-1}) [15]. Mercury concentration in suspended sediment was 2083 ng·g^{-1} of dry weight for suspension obtained from seawater and 290 ng·g^{-1} of dry weight for suspension from sea foam. The accumulation coefficient of mercury by suspended matter (K_{ss}) for seawater was 3.8·10^4, and for foam – 1.5·10^3. Such a distribution of mercury in sea foam and water, as well as K_{ss} values obtained, may indicate a high concentrating ability of marine sedimentary suspensions and their high significance in self-purification of marine area.

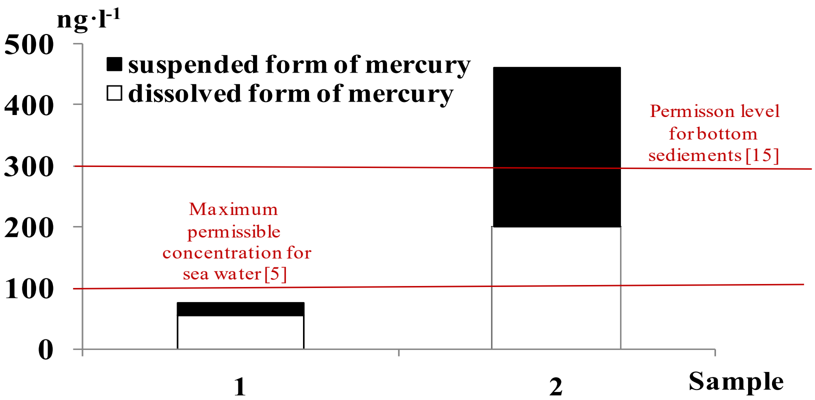

Fig. 7. Sample 1 – concentration of total mercury in seawater, ng·L^{-1}; sample 2 – concentration of total mercury in sea foam, ng·L^{-1}

Conclusion. In the Black Sea, dissolved form of mercury predominates regardless of the season, with a variation in its percentage in the range of 66.3–85.8 % of total mercury concentration. The accumulation coefficient of mercury by suspended matter varied from n·10^3 to n·10^7; at its values > 10^6, almost all mercury was accumulated by suspensions. At low mercury concentrations in water, the concentrating ability of suspensions, due to relatively high K_{ss} values, is a significant factor in sedimentary self-purification of water, but its effect decreases with increasing water pollution by mercury. Concentration of suspended form of mercury makes a significant contribution to total mercury concentration, especially in sea foam. The redistribution of sedimentary suspended matter into foam can serve as a source of mercury remobilization in seawater. Therefore, the limits of changes in the accumulation coefficient can be decisive indicators in self-purification of marine environment. They can be used to solve the problems of limiting mercury emissions to the Black Sea, which can help in the timely identification of possible environmental hazard.

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КОНЦЕНТРИРОВАНИЕ РТУТИ ВО ВЗВЕШЕННОМ ВЕЩЕСТВЕ ПЕНЫ И ВОДЫ ЧЁРНОГО МОРЯ*

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Способность взвешенного вещества концентрировать ртуть может быть превалирующим фактором в очищении водной толщи Чёрного моря. В результате седиментации взвешенные частицы выносят загрязнения из поверхностного слоя воды и в итоге могут депонировать их в донных осадках, участвуя таким образом в процессах самоочищения морской акватории. Взвешенное вещество как дисперсная фаза водной среды, рассматриваемой в качестве гетерогенной дисперсной системы, может быть более насыщено ртутью, чем сама вода как дисперсионная среда. В данной работе определён вклад растворённой и взвешенной форм ртути в её общее содержание и оценена концентрирующая способность взвешенного вещества в отношении ртути, обуславливающая биогеохимическое самоочищение вод от ртути. Все пробы воды разделяли на фильтрат и взвесь путём их фильтрации через нуклеопоровые фильтры с диаметром пор 0,45 мкм. Измерения содержания ртути проводили на анализаторе «Хиранума-1» методом атомно-абсорбционной спектрофотометрии. Концентрацию растворённой ртути в воде определяли в пересчёте на литр, а во взвешенном веществе — на литр и на грамм сухой массы. Выявлено превалирование растворённой формы ртути независимо от сезона года с варьированием её процентного содержания в диапазоне 66,3–85,8 % от общей (суммарной) концентрации ртути. Средняя концентрация взвешенной формы составила 14,2–33,7 % от её общего содержания. При этом значения концентрации взвешенного вещества ($m_{взв}$) варьировали от 0,1 до 15,0 мг·л$^{-1}$ за весь исследованный период, а коэффициент накопления ртути взвешенным веществом ($Кн_{взв}$) изменялся в диапазоне от n·10^3 до n·10^7. Определён значительный вклад

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взвешенной формы ртути в её общее содержание в морской пене, образованной в штормовую погоду. Так, при концентрации взвешенного вещества в морской воде 9,6 мг·л$^{-1}$ концентрация растворённой формы ртути имела значение 55 нг·л$^{-1}$, а взвешенной — 20 нг·л$^{-1}$. В морской пене концентрация взвешенного осадочного вещества составила 895,2 мг·л$^{-1}$, а концентрация ртути достигла 200 нг·л$^{-1}$ в растворённой форме и 260 нг·л$^{-1}$ — во взвешенной. Содержание общей (суммарной) ртути в морской пене при этом превышало предельно допустимую концентрацию (100 нг·л$^{-1}$) для морской воды. В данном случае $Кн_{взв}$ для морской воды был равен 3,8·10^4, а для пены — 1,5·10^3. Такое распределение ртути в морской взвеси, пене и воде, а также полученные значения коэффициента накопления свидетельствуют о большой важности взвешенного вещества в самоочищении морской акватории. При низком содержании ртути в воде концентрирующая способность взвешенного вещества, характеризуемая относительно высокими значениями его коэффициента накопления ртути, становится весьма значимым фактором в седиментационном самоочищении вод от ртути, однако при повышении загрязнения вод ртутью влияние этого фактора снижается.

Ключевые слова: ртуть, взвешенное вещество, морская пена, Чёрное море