

**SUSPENDED PARTICULATE MATTER  
AS A BIOCHEMICAL BARRIER TO HEAVY METALS  
IN MARINE FARM AREAS  
(SEVASTOPOL, THE BLACK SEA)**

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For recreational zones and marine farm areas, the investigation of chemical fluxes in coastal marine areas is of certain relevance. To study the role of suspended particulate matter in formation of biogeochemical barriers in marine farm areas, a method was proposed for estimating the fluxes of sedimentary self-purification of water from heavy metals (Co, Ni, Cu, Zn, Mo, Cd, and Pb) and arsenic (As). Based on the literature data on radioisotope dating of bottom sediments and the sedimentation rate, as well as considering our own estimates of the specific gravity of suspended particulate matter in water and concentrations of dissolved and suspended forms of heavy metals and As in the marine environment, the fluxes of biogeochemical self-purification of a marine farm water area from these trace elements were determined. As shown, the proposed methodological base is applicable for ecological regulation of coastal recreational zones in terms of pollution by trace elements.

**Keywords:** Black Sea, pollution, trace elements, self-purification, ecological regulation, suspended particulate matter, aquaculture

Industrial and recreational activities off the Crimean coast result in an increased entry of pollutants, *inter alia* heavy metals, into the marine environment. Suspended particulate matter (hereinafter SPM) is one of the indicators characterizing the quality of coastal marine water. Depending on its sources, SPM is divided into autochthonous and allochthonous. Allochthonous SPM enters the seas with coastal runoff due to coastal abrasion, resuspension of bottom sediments, river and industrial discharges, atmospheric precipitation, *etc.* As a result, various compounds, both nutrients and pollutants, including heavy metals, enter the marine environment. Biological processes, such as phyto- and zooplankton functioning (formation, destruction, and biochemical transformation of the organic component of suspension), play an important role in the formation of autochthonous SPM (Kukushkin & Parkhomenko, 2021). In the open sea areas, autochthonous SPM prevails, while near estuarine zones, allochthonous SPM predominates. In the coastal sea areas, which are not adjacent to estuarine zones of the rivers but are subject to coastal runoff of terrigenous SPM, waters are characterized by increased trophicity and production of biogenic (autochthonous) material (Gulin *et al.*, 2013). In this publication, the researchers showed that autochthonous SPM prevails at the outer roadstead of Sevastopol, and it indicates a significant role

of biotic processes in the transport of SPM there. The marine farm for cultivating bivalves certainly contributes to SPM formation in this water area. Since SPM is the primary trophic link in aquatic ecosystems, mariculture species interact with the sources of their constitutive, energy, and mineral nutrition contained in SPM, as well as with heavy metals, which can be both trace elements essential for metabolism and toxic pollutants of the marine environment.

According to modern views (Ergül et al., 2008 ; Stetsiuk, 2020 ; Turner & Millward, 2002), the living and abiogenic components of SPM concentrate chemicals of various biological significance, both in sorption and metabolic ways, with very high accumulation factors due to a large specific surface of fine-grained particles. Because of their negative buoyancy, SPM becomes the source of sedimentation processes and causes conservative chemical pollutants associated with SPM to deposit in bottom sediments. As a result, in marine farm areas, biogeochemical barriers may be formed affecting the chemical composition and self-purification of water from pollution.

Between SPM and metals in solution, adsorption/desorption and (co)precipitation occur; therefore, the ratios of trace elements in SPM and in dissolved fraction may vary depending on SPM composition, as well as on hydrological and hydrochemical parameters of seawater (Salomons & Förstner, 1984).

As known, heavy metals in water bodies do not degrade, unlike organic pollutants; instead, heavy metals are redistributed over the aquatic ecosystem components. At the same time, SPM has the greatest concentrating ability in relation to metals (due to its large specific surface area). In the process of sedimentation, suspended particles deposit pollutants in the bottom sediments. This allows considering SPM sedimentation as the main biogeochemical mechanism of self-purification of seawater (Bufetova, 2022 ; Egorov, 2019 ; Matishov et al., 2017).

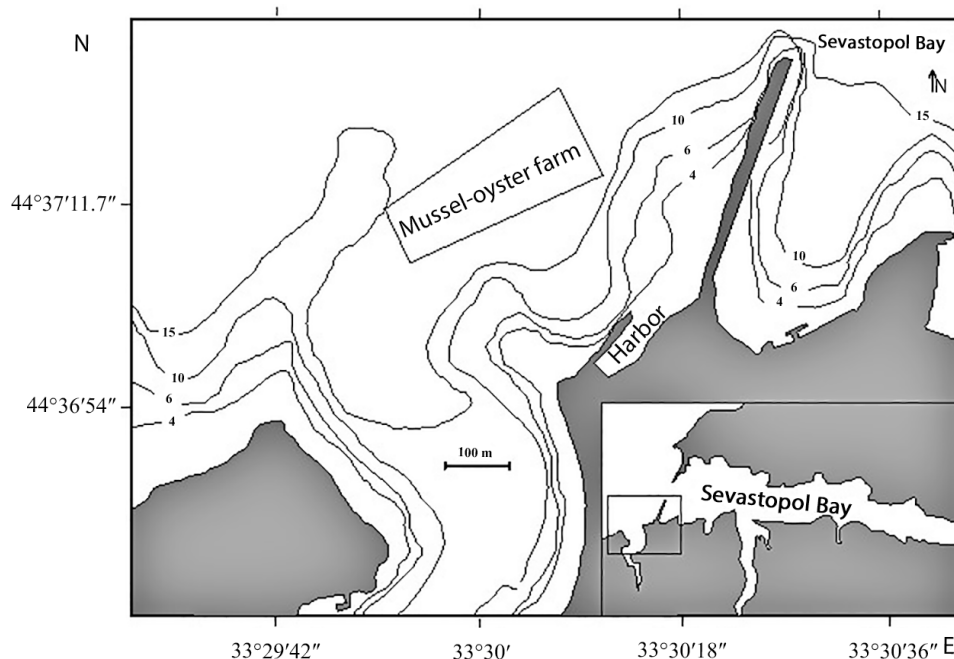
The toxicological hazard of heavy metals and arsenic is normalized by sanitary and hygienic criteria. According to current regulations, water quality is estimated by maximum permissible concentrations (hereinafter MPC) for pollutants dissolved in water (Normativy kachestva vody, 2016). This diagnostic criterion shows threshold concentrations for population. Importantly, content of heavy metals and arsenic in water is formed by pools of dissolved and suspended forms of trace elements. Pools of suspended forms of metals are a biogeochemical barrier. This is of particular relevance in marine farm areas since such a barrier in ecotoxicological terms protects coastal waters and farm products from pollution. In this paper, a methodology for estimating the fluxes of biochemical self-purification of water from heavy metals and arsenic is proposed to study the role of SPM in the formation of biogeochemical barriers in marine farm areas. The method can further be used in the development of criteria for assessing the maximum permissible fluxes of water pollution in marine farm areas, taking into account sanitary and hygienic standards.

## MATERIAL AND METHODS

**Sampling area.** The sampling was carried out at the outer roadstead of the Sevastopol Bay (44°37'13.4"N, 33°30'13.6"E) in 2020 (Fig. 1). The study area was chosen due to its proximity to the coastline (700 m) and location of a marine farm for cultivating mussels and oysters.

The water in this area is classified as mesotrophic; values of the water eutrophication index (E-TRIX) range from 1.63 to 4.33 (Kuftarkova et al., 2006). The mean depth in the marine farm area is 22 m. The long-term average temperature in February is +8 °C; salinity varies within 17.2–18.1 ‰, with temperature and salinity being uniform from the surface down to the bottom. The long-term average

oxygen concentration in water is maximum in winter ( $7.3\text{--}7.7\text{ mL}\cdot\text{L}^{-1}$ ). The content of phosphates does not exceed  $0.37\ \mu\text{M}$ . The concentration of nitrates in surface waters in winter is on average higher ( $0.5\text{--}7.5\ \mu\text{M}$ ) than in summer due to the assimilation by fast-growing phytoplankton (Kapranov et al., 2020). As shown, the formation of the hydrochemical regime of the studied water area is affected by polluted water of the Sevastopol Bay (discharge of the Chernaya River) and Karantinnaya Bay (storm sewer wastewater), emergency discharge of domestic wastewater (up to 3 thousand  $\text{m}^3$  of untreated wastewater *per day*), and storm runoff (Ivanov et al., 2002 ; Nemirovsky & Eremin, 2003).



**Fig. 1.** Map of the study water area and sampling site

**Sampling.** In February 2020, water and SPM were sampled simultaneously to measure trace elements in them. In total, 50 L of seawater were sampled for analysis from the surface. The volume of samples was determined taking into account the expected concentration of SPM, so that the total mass of the collected SPM was 10–50 mg dry weight (hereinafter DW). SPM samples were concentrated using a 6-section vacuum filtration device (at a residual pressure of 0.4–0.6 atm) through the FMNTs-0.45 membrane filters. In total, 6 SPM samples and 6 water samples were treated.

**Measurement of heavy metal content in seawater and SPM.** Heavy metals and arsenic (hereinafter trace elements, TE) were isolated from seawater by the extraction concentration technique according to the guiding document RD 52.10.243-92 (Zolotov & Kuzmin, 1971). The technique consists in the extraction of complexes of the studied elements with carbon tetrachloride using sodium diethyldithiocarbamate (Na-DDC) as a chelating agent, which is followed by the destruction of the complexes with concentrated nitric acid and re-extraction of the elements in an aqueous solution.

SPM concentration was determined by the gravimetric method (Vityuk, 1983). Specifically, the FMNTs-0.45 membrane filters made of cellulose nitrate were kept for 30 min in a drying chamber at a temperature of  $+60\text{ }^{\circ}\text{C}$ ; then, those were kept for 2 h in a desiccator with a moisture absorber ( $\text{CaCl}_2$ ). The weighing was carried out on an analytical scale with an accuracy of 0.1 mg.

After passing a certain volume of seawater through the filters (6 filters *per* sample) under vacuum (at a residual pressure of 0.4–0.6 atm), the filters were washed with deionized water, dried in the same way as described above, and weighed again. SPM concentration was calculated based on the difference between the weights of the filters before and after filtration, taking into account the volume of water passed, and expressed in terms of  $\text{mg}\cdot\text{L}^{-1}$  or  $\text{mg}\cdot\text{m}^{-3}$ .

TE were isolated from the SPM samples by acid mineralization in accordance with the environmental regulatory document PND F 16.2.2:2.3.71-201. The mineralization was performed in a mixture of nitric acid and hydrogen peroxide heated on an electric stove; the filters were completely dissolved, and insoluble residues of SPM were separated by filtration through “Sinyaya lenta” filter papers. In parallel, a blank measurement was carried out with clean filters, using the same amounts of reagents as for the SPM samples.

TE in re-extracts from seawater and mineralized SPM were measured on the basis of IBSS core facility “Spectrometry and Chromatography” by inductively coupled plasma mass spectrometry using PlasmaQuant MS Elite mass spectrometer (Analytik Jena AG, Germany) in accordance with the international technical standard GOST R 56219-2014 and the operating manual for PlasmaQuant MS instruments. The spectrometer was calibrated for the measured elements using an IPC special multi-element standard solution IV (28 elements) (Inorganic Ventures, USA). The error in measurements of TE in seawater was no more than 15 %; in SPM, no more than 10 %.

**Equations for assessing the fluxes of self-purification of water from trace elements *via* sedimentation.** Accumulation factors (AF) of TE by SPM were calculated by the formula:

$$AF = 1,000 \times C_{\text{pm}}/C_{\text{d}}, \quad (1)$$

where  $C_{\text{pm}}$  is content of TE in SPM ( $\mu\text{g}\cdot\text{g}^{-1}$  DW);

$C_{\text{d}}$  is content of TE dissolved in seawater ( $\text{mg}\cdot\text{L}^{-1}$ ).

A series of equations is proposed to evaluate the role of SPM in the formation of biogeochemical barriers against TE in coastal water areas. Theoretical analysis of the obtained results was carried out taking into account the recent views on the radioisotope and chemical homeostasis of marine ecosystems (Egorov, 2019).

The pool of TE in SPM ( $P_{\text{pm}}$ ) under  $1 \text{ m}^2$  of the farm in the 22-m water column ( $\mu\text{g}\cdot\text{m}^{-2}$ ) was calculated as follows:

$$P_{\text{pm}} = C_{\text{pm}} \times 22 \times 1,000. \quad (2)$$

The total pool of TE in seawater and SPM ( $P_{\text{d+pm}}$ ) under  $1 \text{ m}^2$  of the farm in the 22-m water column ( $\mu\text{g}\cdot\text{m}^{-2}$ ) was calculated as follows:

$$P_{\text{d+pm}} = (C_{\text{d}} + C_{\text{pm}}) \times 22 \times 1,000, \quad (3)$$

where  $C_{\text{pm}}$  is concentration of TE in SPM ( $\mu\text{g}\cdot\text{L}^{-1}$ ).

According to the data in (Gulin et al., 2013), the sedimentation flux ( $F_{\text{sed}}$ ) in the study area, calculated using the radioisotope dating method of bottom sediments, was equal to  $664 \text{ g DW}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$ , or  $1.82 \text{ g DW}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ . Daily sedimentation flux of TE ( $V_{\text{sed/day}}$ ) ( $\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ ), or the flux of self-purification of water, was calculated by the formula:

$$V_{\text{sed/day}} = F_{\text{sed}} \times C_{\text{pm}}, \quad (4)$$

where  $F_{\text{sed}}$  is sedimentation flux ( $\text{g DW} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ ).

TE turnover ( $T_{\text{pt}}$ ) (days) due to sedimentation processes in the 22-m layer was estimated using the following equation:

$$T_{\text{pt}} = P_{\text{d+pm}} / V_{\text{sed/day}}. \quad (5)$$

The time constant ( $p$ ) ( $\text{year}^{-1}$ ) was calculated as follows:

$$p = 365 / T_{\text{pt}}. \quad (6)$$

Annual sedimentation flux ( $P_{\text{ad}}$ ) of TE into bottom sediments ( $\text{kg} \cdot \text{ha}^{-1}$ ) was determined by the formula:

$$P_{\text{ad}} = V_{\text{sed/day}} \times 365. \quad (7)$$

Also, TE content in SPM ( $N$ ) ( $\mu\text{g} \cdot \text{g}^{-1} \text{DW}$ ) was calculated:

$$N = C_{\text{pm}} / m_{\text{pm}}, \quad (8)$$

where  $m_{\text{pm}}$  is SPM mass in seawater ( $\text{mg} \cdot \text{L}^{-1}$ ).

**Statistical analysis.** All results in the tables are presented as (mean  $\pm$  standard deviation). The significance level was set as  $p < 0.05$ .

## RESULTS

**Content of trace elements in water and SPM.** It was determined that in February 2020, SPM concentration in the study area amounted to  $0.72 \text{ mg DW} \cdot \text{L}^{-1}$ . The results of measurements of TE concentrations in surface waters and in SPM within the analyzed area are given in Table 1.

**Table 1.** Trace element concentrations in the marine farm water (Sevastopol, February 2020)

TE	Trace element concentration (mean $\pm$ SD)					N (mean $\pm$ SD), $\mu\text{g} \cdot \text{g}^{-1} \text{DW}$	MPC for fishery basins, $\mu\text{g} \cdot \text{L}^{-1}$
	$C_{\text{d+pm}}$ , $\mu\text{g} \cdot \text{L}^{-1}$ (% of MPC)	$C_{\text{d}}$ (dissolved), $\mu\text{g} \cdot \text{L}^{-1}$	% of total	$C_{\text{pm}}$ (SPM), $\mu\text{g} \cdot \text{L}^{-1}$	% of total		
1	2	3	4	5	6	7	8
Co	0.024 (0.4)	$0.019 \pm 0.001$	79.2	$0.005 \pm 0.001$	20.8	$6.93 \pm 0.07$	5
Ni	0.940 (8.7)	$0.866 \pm 0.023$	92.4	$0.071 \pm 0.001$	7.6	$99.76 \pm 0.94$	10
Cu	2.506 (45.0)	$2.248 \pm 0.063$	89.7	$0.258 \pm 0.003$	10.3	$358.18 \pm 4.03$	5
Zn	22.477 (42.7)	$21.374 \pm 0.680$	95.1	$1.103 \pm 0.018$	4.9	$1,532.40 \pm 25.76$	50
As	0.073 (0.3)	$0.032 \pm 0.005$	44.1	$0.041 \pm 0.0004$	55.9	$56.37 \pm 0.52$	10
Mo	1.985 (198.5)*	$1.982 \pm 0.071$	99.8	$0.003 \pm 0.0001$	0.2	$4.44 \pm 0.11$	1
Cd	0.029 (0.3)	$0.026 \pm 0.001$	90.0	$0.003 \pm 0.0005$	10.0	$4.01 \pm 0.07$	10
Pb	0.555 (0.5)	$0.481 \pm 0.012$	86.6	$0.074 \pm 0.001$	13.4	$103.27 \pm 1.87$	10

**Note:** SD denotes standard deviation; \* denotes a value exceeding threshold limit value (MPC).

The data in Table 1 indicate that the ecotoxicological situation in February 2020, concerning water pollution in the marine farm area, can be regarded favorable by all TE (only a certain exceedance of MPC by molybdenum was registered). Importantly, the Russian regulations (*Normativy kachestva vody*, 2016) establish values mostly for freshwater economic objects; only for several TE, the MPC in marine water are additionally provided. For molybdenum, the MPC in marine water is not given.

In the column 2 of Table 1, the values in parentheses are TE concentrations compared to their MPC. Columns 4 and 6 show the ratios of dissolved and suspended forms in the total TE content in seawater. According to the data in Table 1, the total TE concentration in water ranged 0.024–22.477  $\mu\text{g}\cdot\text{L}^{-1}$ . The values for dissolved forms were within 0.019–21.374  $\mu\text{g}\cdot\text{L}^{-1}$ ; for suspended forms, within 0.003–1.103  $\mu\text{g}\cdot\text{L}^{-1}$ . Analyzed TE can be arranged in a sequence of their decreasing level in water as follows: Zn > Cu > Mo > Ni > Pb > As > Cd > Co. For SPM, the sequence was as follows: Zn > Cu > Pb > Ni > As > Co > Mo > Cd.

The data in Table 1 and the available literature material on the sedimentation rate of total (allochthonous and autochthonous) biogenic matter in the study area (Gulin et al., 2013) made it possible to propose a methodology for assessing the role of SPM in the formation of biogeochemical barriers in coastal waters (for example, in the marine farm area), based on the series of equations (2–7). The results of assessing the biogeochemical characteristics of the analyzed water area are summarized in Table 2.

**Table 2.** Biogeochemical characteristics of the turnover of trace elements in suspended particulate matter in the marine farm water area (Sevastopol, February 2020)

TE	AF, $\times 10^4$ DW	$P_{d+pm}$ , $\mu\text{g}\cdot\text{m}^{-2}$	$P_{pm}$ , $\mu\text{g}\cdot\text{m}^{-2}$	$V_{\text{sed/day}}$ , $\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	Parameters of the sedimentation TE turnover in the 22-m water column		$P_{ad}$ , $\text{kg}\cdot\text{ha}^{-1}$
					$T_{pt}$ , day	$P_{ad}$ , $\text{kg}\cdot\text{ha}^{-1}$	
1	2	3	4	5	6	7	8
Co	$30 \pm 1$	$5.3 \cdot 10^2$	$1.0 \cdot 10^2$	12.6	42.0	8.69	0.045
Ni	$10 \pm 0.3$	$206.1 \cdot 10^2$	$15.6 \cdot 10^2$	181.6	113.5	3.22	0.663
Cu	$16 \pm 0.5$	$547.8 \cdot 10^2$	$56.8 \cdot 10^2$	651.9	84.8	4.30	2.379
Zn	$7 \pm 0.2$	$4,944.9 \cdot 10^2$	$242.7 \cdot 10^2$	2,788.9	177.4	2.06	10.180
As	$180 \pm 8$	$16.1 \cdot 10^2$	$9.0 \cdot 10^2$	102.6	15.8	23.10	0.374
Mo	$0.20 \pm 0.02$	$436.7 \cdot 10^2$	$0.7 \cdot 10^2$	8.1	5,404.7	0.07	0.029
Cd	$15 \pm 0.5$	$547.8 \cdot 10^2$	$6.4 \cdot 10^2$	7.3	87.7	4.16	0.027
Pb	$0.020 \pm 0.001$	$4,944.9 \cdot 10^2$	$122.1 \cdot 10^2$	187.9	65.0	5.62	0.684

Calculations showed that the AF values ranged  $0.02 \cdot 10^4$  to  $180.00 \cdot 10^4$  (column 2 in Table 2). The total pool of TE in water ( $P_{d+pm}$ ) varied from  $5.3 \cdot 10^2$  to  $4,944.9 \cdot 10^2$   $\mu\text{g}\cdot\text{m}^{-2}$  (column 3); out of it, TE content in SPM was 0.2–55.9 % (column 6 in Table 1). The flux of self-purification of water *via* sedimentation ( $V_{\text{sed/day}}$ ) ranged within  $7.30$ – $2,788.97$   $\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$  (column 5 in Table 2). The TE turnover in the marine environment due to their concentration and elimination through the sorption and sedimentation processes mostly had a time scale from days to months or seasons. The only TE with the turnover



on an annual time scale was Mo (column 6). Based on these data, we obtained the following sequence in ascending order for  $T_{pt}$  (days): As < Co < Pb < Cd < Cu < Ni < Zn < Mo. Arsenic is the TE with the shortest SPM cycle (within 15 days); Co, Pb, Cd, and Cu are deposited with SPM within 1.5–2.5 months; a complete turnover of Ni and Zn in the cycle takes 3–6 months; and molybdenum has the slowest cycle (more than 14 years).

In column 7 of Table 2, we provide the estimates for the time constants of sedimentation exchange of TE content pools in the water layer 0–22 m. They showed: as a result of biogeochemical processes, TE content in this layer can be replaced 0.07–23.10 times a year. Taking into account this circumstance, a solution can be provided to the problem of ecological regulation of anthropogenic load on marine water areas, including regulation of the maximum permissible fluxes of TE entering waters of recreational zones. According to the results of calculations given in column 8 of Table 2, with the chemical composition of waters remaining at a stationary level in February 2020, the rate of sedimentation and deposition of various TE in deep layers of bottom sediments in each hectare of the studied farm water area ranged 0.027 to 10.180 kg·year<sup>-1</sup>.

## DISCUSSION

**On SPM composition.** Comparison of the results obtained in this work with literature material showed that our data on SPM are consistent with previously published information. Specifically, according to (Ryabushko et al., 2014), SPM content in the study area throughout the year varied 0.3 to 1.1 mg·L<sup>-1</sup>; in our work, the value was 0.72 mg·L<sup>-1</sup>.

Plankton is known to play an important role in the formation of autochthonous SPM in the surface layer of the Black Sea. Literature data on the phytoplankton ratio in SPM composition are extremely scarce. Researchers tend to show the ratio of mineral and organic components of SPM. For the Black Sea, the level of organic compounds in SPM ranges from 14 to 88 % (Vityuk, 1983). The ratio of detritus in SPM is significant as well. In the open Black Sea, at the outer roadstead of Sevastopol, SPM mainly consisted of autochthonous suspension, depending on the plankton composition (Gulin et al., 2013 ; Kukushkin & Parkhomenko, 2021); this indicates a notable role of biotic processes in the SPM transport into this area. In phytoplankton during our study, diatoms prevailed (*Pseudo-nitzschia* spp. and *Skeletonema costatum* (Greville) Cleve, 1873), as well as a coccolithophorid *Emiliania huxleyi* (Lohmann) W. W. Hay & H. Mohler, 1967. Wet biomass of phytoplankton in February 2020 in the marine farm area was about 0.1 mg·L<sup>-1</sup> (Pospelova & Priimak, 2021). Assuming that phytoplankton DW averages 10 % of its wet biomass (Studenikina et al., 1999), we can conclude that dry biomass of microalgae in winter was 0.01 mg·L<sup>-1</sup>, or 1.4 % of SPM mass. Apparently, the contribution of phytoplankton to SPM during this study was insignificant. However, it should be taken into account that small-celled species of microalgae prevailed. Those form a large surface area; therefore, they can play a key role in TE concentration from water.

In marine farm areas, cultivated molluscs can contribute to the formation of the chemical composition of seawater and SPM – due to peculiarities of the sorption and trophodynamic processes in them. Molluscs can release both dissolved and suspended organic and mineral substances into the farm water area (Lomakin et al., 2007 ; Pospelova et al., 2018).

An allochthonous contribution to SPM composition in the upper layer of the studied water area can be made by the water runoff of the Chernaya River (through the Sevastopol Bay), two emergency episodically operating wastewater outlets in the immediate vicinity of the farm, and wastewater from storm

sewers entering directly the farm water area, *inter alia* from the Karantinnaya Bay (Ivanov et al., 2002 ; Nemirovsky & Eremin, 2003).

**Content of dissolved and suspended forms of trace elements in water.** According to different researchers, the values of concentrations of the studied TE for the Black Sea vary widely (Table 3). Comparison of our results with those available in literature showed as follows: the values of total concentration of Ni, Cu, Zn, Cd, and Pb in the marine farm area are within the ranges of variation of clarkes of these TE in the Black Sea, whereas content of Co and As in this work is lower than mean values for the Black Sea (Table 3). Concentrations of all TE, except for Zn, in SPM are comparable to the average data for the Black Sea.

**Table 3.** Variability ranges of trace element concentrations in the Black Sea, trace element content in suspended particulate matter of the Chernaya River, and clarkes in the Earth's crust

TE	TE concentration in the Black Sea			Clarkes in the Earth's crust, $\mu\text{g}\cdot\text{g}^{-1}$ (Kasimov & Vlasov, 2015)	TE concentration in SPM at the Chernaya River mouth, $\mu\text{g}\cdot\text{g}^{-1}$ (Malakhova et al., 2020)
	Total concentration, $\mu\text{g}\cdot\text{L}^{-1}$	Dissolved form, $\mu\text{g}\cdot\text{L}^{-1}$	Suspended form, $\mu\text{g}\cdot\text{L}^{-1}$ (% of total)		
Co	0.6–3.1 <sup>4)</sup>	nd	0.60·10 <sup>-3</sup> ...0.10 <sup>1)</sup>	15	14
Ni	0.4–23 <sup>4)</sup>	0.47–0.70 (max 8.334) <sup>2)</sup>	0.005...0.26 <sup>1)</sup> (0.01) <sup>5)</sup>	50	76
Cu	< 0.01–33 <sup>3)</sup>	0.06–0.51 (max 7.75) <sup>2)</sup>	0.006...0.29 <sup>1)</sup> (0.03) <sup>5)</sup>	27	877
Zn	0.42–108 <sup>3)</sup>	max 54.53 <sup>2)</sup>	0.001...0.91 <sup>1)</sup> (0.25) <sup>5)</sup>	75	1,273
As	0.3–2.5 <sup>4)</sup>	0.83–1.3 <sup>4)</sup>	nd	5.6	14
Mo	0.67–3.74 <sup>6)</sup>	nd	0.3·10 <sup>-3</sup> ...0.07 <sup>1)</sup>	1.1	1
Cd	< 0.01–0.45 <sup>3)</sup>	max 1.69 <sup>2)</sup>	0.001...3.60 <sup>1)</sup>	0.09	5
Pb	< 0.01–2.5 <sup>3)</sup>	0.02–0.04 (max 8.08) <sup>2)</sup>	0.005...0.35 <sup>1)</sup> (0.015) <sup>5)</sup>	17	196

**Note:** <sup>1)</sup>Yiğiterhan et al., 2011; <sup>2)</sup>Boran & Altinok, 2010; <sup>3)</sup>Sevost'yanova et al., 2016; <sup>4)</sup>Mitropolsky et al., 1982; <sup>5)</sup>Patin и Morozov, 1981; <sup>6)</sup>Nägler et al., 2011; nd denotes no data.

The ratio of suspended form of Ni, Cu, Zn, and Pb in the studied water area is higher than the average for the Black Sea. Importantly, the literature data used by us for comparison refer to the 1970–1980s, and the disagreement between the numbers is likely to result from both the difference in the techniques for determining SPM concentration and temporal changes in SPM composition.

Specifically, Cu, Zn, and As are geochemically mobile elements. Cu and Co are largely assimilated by the biota, while As is often associated with SPM. Ni is mostly in the dissolved phase in the marine environment. Pb is characterized by high activity of interaction with living matter (Mitropolsky et al., 2006). Mo is less biologically available in seawater than in freshwater (Howarth et al., 1988), and there is usually 10,000 times more dissolved Mo in seawater than in SPM (Dellwig et al., 2007 ; Howarth & Cole, 1985).

As known, there are no regulations for TE content in SPM of seawater; therefore, for a comparative assessment of the degree of environmental pollution, we used clarkes of chemical elements in the upper continental crust (Kasimov & Vlasov, 2015) (Table 3). The comparison of the obtained



results showed that cadmium content in SPM of the farm was 44 times higher than mean clarkes of this element in the Earth's crust; zinc, 20 times; copper, 13 times; and lead, 6 times (Table 3). Cobalt content was 2 times lower. Apparently, this is due to the effect of both natural and anthropogenic factors. Natural factors include TE supply from bottom sediments, as well as with the waste products of cultivated molluscs and planktonic organisms. Anthropogenic factors will be considered in more detail.

An increase in the concentration of pollutants in the surface waters of the analyzed area is largely due to the effect of the adjacent polluted bays – the Karantinnaya Bay (Ryabushko et al., 2017) and Sevastopol Bay (Orekhova & Varenik, 2018 ; Sovga & Mezentseva, 2019). The Sevastopol Bay – a large semi-enclosed basin of the estuarine type – experiences certain anthropogenic pressure. In turn, its ecological state is affected by coastal sources of pollution, active navigation, and the Chernaya River runoff. At the same time, in the Sevastopol Bay, currents and winds develop which carry pollutants off the bay and contaminate the marine farm area (Kapranov et al., 2020). During our sampling, northeast winds prevailed in the Sevastopol region (<https://weatherarchive.ru/Sevastopol/19-February-2020>), which may evidence for the effect of the bay water on the marine farm area. In parallel with the data of our research, in February 2020, results were obtained on TE content at the Chernaya River mouth (Table 3) (Malakhova et al., 2020). Significant amounts of Cu, Zn, and Pb entered the Sevastopol Bay with SPM from the river. Concentrations of Co, Cu, and Pb in SPM of the river were higher than in SPM of the marine farm area; content of Ni, Zn, and Cd was comparable; and As and Mo were concentrated in SPM of the farm with a greater intensity than in SPM of the river. As known, in the mixing zone of river water and seawater, a geochemical barrier is formed. There, due to hydrological, physicochemical, and biological processes, more than 90 % of SPM of the river runoff is removed from water, and up to 90 % of suspended forms of Mn, Co, Ni, Cu, Zn, As, Cd, and Pb are eliminated from further migration, as well as 10–60 % of dissolved forms (Demina, 2015). So, the Chernaya River water could not make a significant contribution to TE content in the marine farm area.

Usually, in coastal zones, TE of aerosols (dissolved and suspended) are involved in biogeochemical cycles (Duce et al., 1991). The atmospheric component plays a notable role in the entry of pollutants, *inter alia* TE, into the Black Sea. Up to 83 % of Ni, Cu, Zn, Cd, and Pb fluxes into the Black Sea are provided by atmospheric fallout (Gubanov et al., 2004 ; Theodosi et al., 2013). The atmospheric contribution of Ni, Cu, and Pb to the marine environment of the Sevastopol region is comparable to the inflow of river runoff and sometimes even exceeds it (Ovsyanyi et al., 2001). Presumably, high content of suspended forms of Pb (predominantly of technogenic origin) is associated with the formation of aerosols with a high content of lead because of pollution from automobiles and from coal combustion; also, it is associated with industrial effluents. Arsenic is the substance of the first hazard class and is included in the group of chemical elements that must be detected during environmental monitoring, especially in marine farm areas. The results of our work showed that more than 50 % of As is present in water in suspended form. Similar data are published for the Chernaya River water (Malakhova et al., 2020). However, total concentrations of As and Pb in water did not exceed MPC (Table 1). The only heavy metal with content in water of 1.98 MPC was Mo. It is the most abundant transition metal in open sea water – due to the prevalence and low chemical reactivity of the molybdate ion ( $\text{MoO}_4^{2-}$ ). Rivers supply oceans with most of molybdenum, mainly due to the weathering of continental material (Smedley & Kinniburgh, 2017).

**Fluxes of trace elements through SPM.** We assessed (Table 2) the dynamic parameters of the TE absorption by SPM by calculating the duration of biogeochemical cycles of these TE. Even with a lower ratio of metals in SPM, compared with that in the dissolved phase, SPM significantly (from several days to several years) accelerates the TE turnover.

The AF values obtained by us (Table 1) testify to high ability of SPM to concentrate TE. The ratio of metals in suspension (0.2–55.9 % of their total content in the aquatic environment) was lower than in the Sea of Azov (29–95.6 %) (Bufetova, 2022). It can be explained by SPM content in water: in the Black Sea, it is an order of magnitude lower than in the Sea of Azov. An increase in the ratio of metals in SPM with a rise in SPM content was also shown for the Sea of Japan coastal areas (Shulkin, 2007).

TE pool in SPM in the marine farm area ranged from 70  $\mu\text{g}$  to 24 mg *per*  $\text{m}^2$  of the water column; the values were minimum for Mo (70  $\mu\text{g}$ ) and Co (100  $\mu\text{g}$ ) and maximum for Zn (24 mg) and Pb (12 mg). The pool values made it possible to calculate the flux of self-purification of water from TE *via* sedimentation. Similar data on vertical fluxes of most of studied TE were obtained for the southeastern Black Sea (Ergül et al., 2008) and for the Caucasus coast (Denisov & Latun, 2018). The metal turnovers in the Sea of Azov are reported in (Matishov et al., 2017). There, the researchers showed the patterns of heavy metal concentration by bottom sediments; based on those, self-purification of water *via* sedimentation was characterized. In this work, the sedimentation turnover for a heavy metal in the aquatic environment ( $T_{\text{pt}}$ , years) was shown to reflect the time scale of self-purification of water *via* sedimentation. Different duration of biogeochemical cycles for various TE can be explained by the differences in their concentrations in solution and ratios of bioavailable forms (Demina, 2011). The calculation techniques proposed by us showed as follows: in the marine farm area, only due to sedimentation processes, water is purified from significant part of toxic elements during the day, and this undoubtedly makes a notable contribution to the purification of the coastal water area. Taking into account the fact that the marine farm for cultivating filter-feeding molluscs is located there, SPM can be considered as an additional factor for ensuring the safety of aquaculture products.

Our calculation of the sedimentation turnover for TE in the aquatic environment ( $T_{\text{pt}}$ , days) (Table 2) indicates the time scales of the processes of self-purification of the marine farm area *via* sedimentation, as well as the estimates for the time constants of sedimentation exchange of TE content pools in the entire water layer of the studied area. This will help in solving the problem of ecological regulation of anthropogenic pressure on marine water areas, *inter alia* regulation of the maximum permissible fluxes of TE which enter waters of recreational zones. Along with the known data on significant contribution of marine farms for cultivating bivalves to self-purification of coastal waters, another key factor should be taken into account – the participation of SPM in these processes.

**Conclusion.** The article presents assessment of the role of suspended particulate matter in marine farm areas as a biogeochemical barrier that contributes to the process of self-purification of the marine area from trace elements. Based on the obtained data on TE content in seawater and SPM in the marine farm area, the fluxes of their sedimentation turnover were determined; moreover, the method was proposed for assessing the process of self-purification of water from heavy metals (Co, Ni, Cu, Zn, Mo, Cd, and Pb) and arsenic (As) taking into account sanitary and hygienic standards. From a practical point of view, the results obtained can be used in the development of criteria for regulating the maximum permissible fluxes of water pollution into other water areas where marine farms are located.

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**ВЗВЕШЕННОЕ ВЕЩЕСТВО  
КАК БИОГЕОХИМИЧЕСКИЙ БАРЬЕР ДЛЯ ТЯЖЁЛЫХ МЕТАЛЛОВ  
В РАЙОНАХ РАЗМЕЩЕНИЯ МОРСКИХ ФЕРМ  
(СЕВАСТОПОЛЬ, ЧЁРНОЕ МОРЕ)**

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Исследования потоков химических веществ в прибрежных морских акваториях приобретают особенную актуальность в рекреационных районах и в местах размещения морских ферм. Для изучения роли взвешенного вещества в формировании биогеохимических барьеров в районах размещения морских ферм предложен метод оценки потоков седиментационного самоочищения вод от тяжёлых металлов (Co, Ni, Cu, Zn, Mo, Cd и Pb) и мышьяка (As). На основе



литературных данных по радиоизотопным датировкам донных отложений и скорости осадко-накопления, а также на базе собственных оценок удельной массы взвешенного вещества в воде, концентраций растворённой и взвешенной форм тяжёлых металлов и мышьяка в морской среде определены потоки биогеохимического самоочищения акватории размещения морских ферм от тяжёлых металлов и мышьяка. Показано, что предложенная методическая база применима для экологического нормирования прибрежных рекреационных зон по фактору загрязнения тяжёлыми металлами и мышьяком.

**Ключевые слова:** Чёрное море, загрязнение, микроэлементы, самоочищение, экологическая регуляция, взвешенное вещество, аквакультура