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Pollution and Potential Ecological Risk Evaluation of Heavy Metals and Arsenic in Surface Marine Sediments of the Coastal Vostok Bay (Peter the Great Bay, Sea of Japan, Russia)

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ABSTRACT

The pollution and potential toxicity of heavy metals (Cu, Cd, Pb, Zn, Ni and Co) and As in the surface bottom marine sediments of the coastal Vostok Bay in 2015 and 2020 were analyzed. Pollution and ecological risk indices were calculated by comparing the concentrations obtained with background, permissible levels and sediment quality standards (SQGs). Maximum concentrations of Cu (37.64±0.88 µg/g), Cd (0.25±0.03 µg/g), Pb (123.73±5.39 µg/g), Zn (162.58±10.31 µg/g), Ni (29.50±1.01 µg/g), Co (5.00±0.23 µg/g) and As (5.24±0.23 µg/g) were detected in the industrialized area of Gaydamak cove in the samples of 2015. Sediments from this area were characterized by moderate pollution and low level of potential toxicity for marine hydrobionts based on the calculation of mCd and TRI. The general trend of decrease in the content of analyzed pollutants in sediments is noted in 2020. Maximum content of Cu (16.17±0.38µg/g), Cd (0.27±0.03µg/g), Pb (58.62±2.55µg/g), Zn (83.14±5.27µg/g), Ni (5.81±0.2µg/g) and Co (5.00±0.23µg/g) was observed in the area of Gaydamak cove as well as in 2015. Low levels of contamination and no potential toxic effects were noted. The highest concentration of As (14.32±0.83 µg/g) was detected in sediments of Srednyaya Cove. This fact is of particular concern as this cove is part of the Integrated Marine Reserve and is characterized by high biodiversity. The results of this study allow us to conclude that the use of individual and complex indices of pollution and potential ecological risk is an effective tool for assessing the ecological status of bottom sediments.

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INTRODUCTION

The problem of pollution of marine areas, in particular coastal zones, by toxic substances is currently of global nature. Technological and economic development of cities leads to the growth of harbor and industrial areas on the coasts of bays and coves. The functioning of facilities in these industrial areas leads to the active inflow of pollutants into the sea, which has a negative impact on the ecosystems of coastal zones (Buruaem et al., 2013; Manzo et al., 2022).

Heavy metals **(HM)** are among the group of priority environmental pollutants due to their high toxicity, resistance to biodegradation and ability to bioaccumulate. HM levels are monitored in all natural environments (Wilbers et al., 2014). HM enter the marine environment as a result of natural processes (chemical and physical weathering of rocks) and anthropogenic activities

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(such as sewage and storm water runoff from agricultural and mining facilities, landfills for recycling and utilization of industrial and domestic wastes) (Ardila et al., 2022; Shang et al., 2023; Ardila et al., 2024)**.** Toxic elements accumulate in bottom sediments through deposition, adsorption, and incorporation mechanisms in biological material (Szefer et al., 1996). Metals from bottom sediments can enter the bottom layer of water through a variety of processes of a physical, chemical, or biological nature and cause secondary contamination (Petukhov et al., 2023). Bottom sediments are an excellent integrator of chronic changes in marine ecosystems, particularly in their chemical composition. Metals such as Pb, Cd and Ni are indicators of the technogenic pressures associated with the activities of various industrial enterprises. The distribution of these metals is particularly influenced by atmospheric transport. Zn and Cu can enter the marine environment not only from industrial sources but also from municipal and domestic wastewater (Khristoforova et al., 2004). Information on the concentration of these pollutants provides an overview of the ecological situation in the aquatic environment (Choi et al., 2010; Luo et al., 2021).

Chemical elements such as Mn, Fe, Cu, Zn, Co, Cr and others are biologically active and take part in physiological processes in organisms. At concentrations above specific thresholds, the elements become toxic, while Cd, Pb and Hg even at low concentrations have pronounced toxic and carcinogenic properties (Abdu et al., 2017; Jafarabadi et al., 2017; Huang et al., 2018; Vezzone et al., 2019; Cunha et al., 2021; Liao et al., 2022). HM contamination of sediments is a risk factors for hydrobionts associated with this substrate, which is manifested in the accumulation of these toxicants in the organs of aquatic organisms, which in turn leads to the development of diseases and death. High toxicity of individual HM and their mixtures for various members of marine hydrobionts has been proven in numerous studies (Chiarelli & Roccheri, 2014; Naz et al., 2023; Jeong et al., 2023).

Sediment quality standards (SQGs) are a tool to assess the potential toxicity of pollutants in sediments to benthic organisms. The basic principle of these regulations is that they can be used as a substitute for toxic effect studies using biological systems. The use of these guidelines in combination with field studies has shown that SQGs, on par with direct toxicological tests, indicate the possibility of adverse acute exposure or no exposure at all (Wenning & Ingersoll, 2004). The use of SQGs protocols allowed the development of integrated sediment toxicity indices to assess the overall benthic hazard level of analyzed pollutants. Assessment of the level of marine sediment pollution is carried out by comparing the concentrations of heavy metals with background values for the study area and international normative values (Warmer & van Dokkum, 2002).

Numerous studies of hydrologic, hydrochemical and microbiological parameters of the waters of Vostok Bay (Peter the Great Bay, Sea of Japan) indicate the increasing influence of anthropogenic pressure on the area (Barysheva et al, 2019; Grigorieva et al, 2020; Khristoforova et al, 2020; 2023). Monitoring works are carried out in the Vostok Bay to assess the accumulation of HM in green and brown algae (Kozhenkova et al., 2006; Chernova and Kozhenkova, 2016; Chernova and Kozhenkova, 2020) and mollusks (Kartavtsev et al., 2001; Podgurskaya & Kavun, 2005) for many years. It is significant to note that the analysis of the HM concentration in the bottom sediments of this bay is described only in few study (Khristoforova et al., 2004; Shulkin, 2004). The aim of this research is to provide an integrated assessment of the current level of contamination of bottom sediments of Vostok Bay with HM and arsenic, and determine of the potential level of toxicity.

MATERIALS & METHODS

Description of the study area

Vostok Bay is a second-order bay, a part of the Peter the Great Bay (Sea of Japan) with a water mirror area of 35.2 km^2. It is a shallow area, not separated by a threshold or constriction from the sea. The coastal zone of the west coast is deeper (with depths up to 10 m), compared to the east and apex coast (maximum depths do not exceed 3-5 m). The low and sandy coast of the northern part of the bay is indented by the mouths of the Volchanka and Litovka rivers (catchment area 197 km^2 and 446 km^2 respectively). The bottom sediments of Vostok Bay are mosaic and their distribution depend on the influence of currents and coastal flow. In the open parts of the bay sands of different fractions are distributed, in the central part muddy sands dominate, in the closed bays – clayey silts are predominant (Gaiko, 2017).

The State Natural Integrated Marine Reserve of regional significance "Vostok Bay" occupies a significant part of the water area (18.2 km^2). It includes the territories of Srednyaya, Vostok, Tikhaya Zavod' and Litovka coves. The majority of phylogenetic groups recorded in the waters of Peter the Great Bay inhabit the relatively small area of Vostok Bay. The unique biological diversity of this area is associated with a variety of habitat conditions: many types of coasts, bottoms, reliefs, landscapes, marine currents, a wide range of water temperature, (from -1.8 to +27 ^{o}C) salinity (from 10 ‰ near the river mouths to 32 ‰ in the open part of the bay) and balanced eutrophication of Vostok Bay (Grigoryeva & Kashenko, 2010; Dolganov & Tyrin, 2014).

The coasts of the Bay are indented by many coves. The most significant of them in the west are Gaydamak and Srednyaya coves. The large industrial cluster located on the coast of Gaydamak cove, which includes the port area, ship repair, fishing and fish processing facilities, makes a significant contribution to the inflow of various types of pollutants, including heavy metals, into the water. The coasts of Srednyaya, Litovka and Antares coves are favorite places of rest for residents of the Russian Far East. The growth of the number of equipped recreation areas, campgrounds, yacht clubs and their operation, especially in the summer , contributes to the pollution of waters and bottom sediments of the bay. The industrial zone and construction base of Rosneft's Eastern Petrochemical Complex is located in Antares cove on the western coast of Vostok Bay (Grigorieva et al., 2020; Khristoforova et al., 2023).

Description of sampling and sample preparation

Bottom sediments were sampled by SCUBA divers in August 2015 and July 2020 from 8 stations located along the western, northern and eastern coasts of Vostok Bay (Fig. 1). The surface layer of sediment, no more than 5 cm deep, was captured using a 10 cm^2 teflon sampler. Three samples were taken from each station and subsequently pooled, frozen and stored at -18 ^{o}C. Samples for physicochemical analyses were defrosted and dried to air-dry condition just before analysis.

Physical and chemical analysis of bottom sediments

The particle size distribution was determined by sieve analysis by sieving a weighted sediment sample through a set of sieves with a pallet in accordance with Interstate standard 12536-2014 (2015). The following sediment fractions were analyzed: silt and clay (<0.1 mm), sand (0.1-2 mm), and gravel (>2 mm). Samples were ground into a powder using a porcelain mortar and pestle. Organic carbon content (TOC) was determined using a modified Tyurin method (Savich et al., 2013) using a sediment fraction with a particle size not exceeding 0.5 mm.

The elements Cu, Cd, Pb, Zn, Ni, Co and As were analyzed using Shimadzu AA-6800 atomic absorption spectrophotometer (Shimadzu Corporation, Japan) with the guidance "Determination of As, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sb, Sn, Zn (acid-soluble forms) in soils and bottom sediments by atomic absorption method" № M-02-902-125-2005 (Novikov, 2022). Determination of Cd, As was performed with an electrothermal atomizer with graphite furnace GFA-6800 (Shimadzu Corporation, Japan). The elements Cu, Pb, Zn Ni, Co were determined in acetylene/air flame. Bottom sediment samples dried at 105 °C were ground and sieved through a capron sieve (mesh size 0.5 mm). Samples weighing 1 g were acid mineralized using nitric and perchloric acids of high purity in the ratio 3:1. Acid-soluble forms of heavy metals were determined in the obtained mineralizate. The accuracy of measurement of element concentration was controlled by analyzing standard samples of metal solutions (GSORM), entered in the State

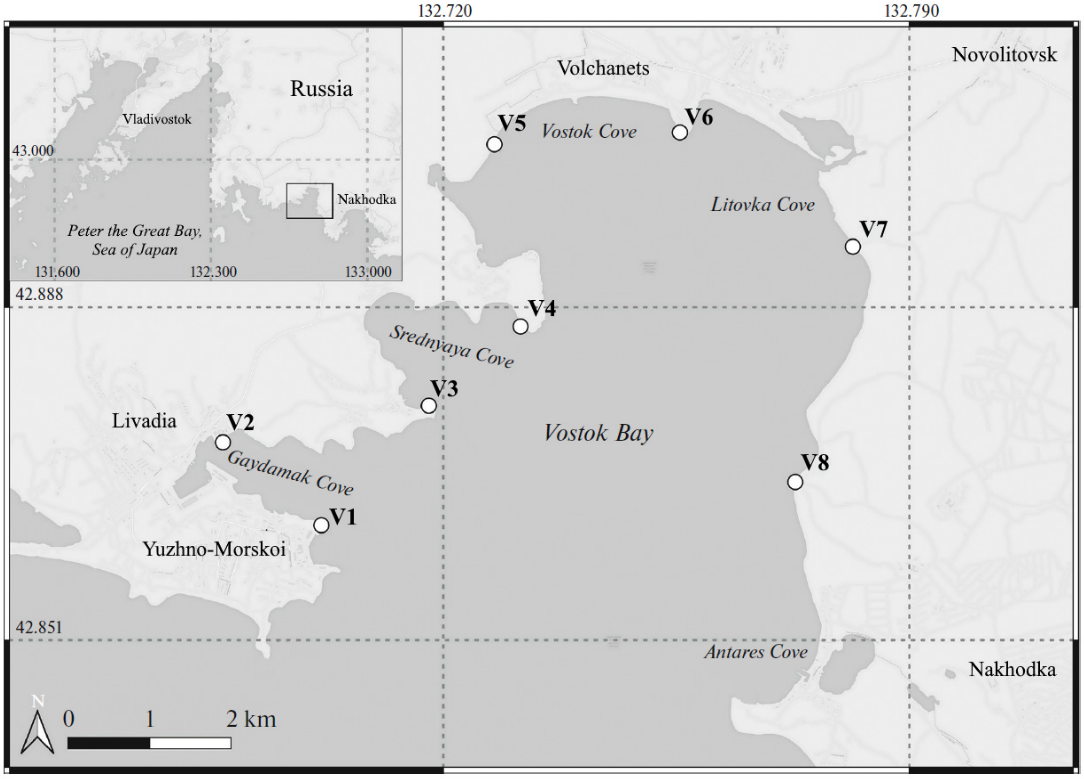


Fig. 1. A schematic map of the area of marine sediments sampling: (1) Cape Chaikovsky; (2) apex of Gaydamak Cove; (3) Cape Pushchin; (4) Cape Pashinnikov; (5) Volchanka River mouth; (6) Cape near the Volchanetskaya Channel; (7) Litovka River mouth; (8) Cape Elizarov

Register of Measurement Devices, contamination of reagents – by means of blank samples.

Calculations of Pollution and Environmental Risk Indices

In order to assess the level of contamination of the analyzed sediments, we calculated the individual enrichment factor (*EF*) as well as the complex modified contamination degree index (*mCd*) (Brady et al., 2014), whose corresponding classifications of values are presented in Table 1. The calculation of these indices is based on the ratio of detected heavy metal concentrations to their background levels presented in Table 2 (Shulkin, 2004; Kovekovdova & Simokon, 2004).

Enrichment factors (*EF*) (Li et al., 2015) are calculated using the following formula:

$$EF = \frac{(C_n / C_{Fe})_{sample}}{(B_n / B_{Fe})_{background}}$$

Where $(C_n / C_{Fe})_{sample}$ – ratio of heavy metals and arsenic to Fe content in the sample, $(B_n / B_{Fe})_{background}$ – ratio of background values of heavy metals and arsenic content to Fe.

Modified contamination degree (*mCd*) index (Brady et al., 2014) was calculated as follows:

$$m\mathrm{C}_d = \frac{\sum_{i=1}^{n} Cf^i}{n}$$

Where Cf^i – the ratio of heavy metal and arsenic concentration in the sample to its background value, n – total quantity of analyzed toxicants.

Table 1. The classification of the indices values

Class	Enrichment factors (*EF*)		Modified hazard quotient (*mHQ*)	
	Value	Enrichment Level	Value	Contamination Degree
1	$EF \leq 1$	no enrichment	$mHQ \leq 0.5$	nil to very low severity of contamination
2	$1 < EF \leq 3$	minor enrichment	$0.5 < mHQ \leq 1.0$	very low severity of contamination
3	$3 < EF \leq 5$	moderate enrichment	$1.0 < mHQ \leq 1.5$	low severity of contamination
4	$5 < EF \leq 10$	moderately severe enrichment	$1.5 < mHQ \leq 2.0$	moderate severity of contamination
5	$10 < EF \leq 25$	severe enrichment	$2.0 < mHQ \leq 2.5$	considerable severity of contamination
6	$25 < EF \leq 50$	very severe enrichment	$2.5 < mHQ \leq 3.0$	high severity of contamination
7	$EF > 50$	extremely severe enrichment	$3.0 < mHQ \leq 3.5$	very high severity of contamination
8			$mHQ > 3.5$	extreme severity of contamination

Class	Modified contamination degree (*mCd*)		Toxic risk index (*TRI*)	
	Value	Contamination Level	Value	Toxic Risk Degree
1	$mCd \leq 1.5$	non to very low degree	$TRI \leq 5$	no toxic risk
2	$1.5 < mCd \leq 2$	low degree	$5 < TRI \leq 10$	low toxic risk
3	$2 < mCd \leq 4$	moderate degree	$10 < TRI \leq 15$	moderate toxic risk
4	$4 < mCd \leq 8$	high degree	$15 < TRI \leq 20$	considerable toxic risk
5	$8 < mCd \leq 16$	very high degree	$TRI > 20$	very high toxic risk
6	$16 < mCd \leq 32$	extremely high degree		
7	$mCd > 32$	ultra high degree		

The degree of toxicity of the studied sediments was determined by calculating the individual modified hazard quotient (*mHQ*) and the composite toxic risk index (*TRI*). The classification of the *mHQ* and *TRI* values in this paper is show in Table 1. These indices are based on the comparison of the obtained concentrations of the studied pollutants with international standard values, namely the SQG sediment quality guidelines (MacDonald et al., 2004) presented in Table 2.

The modified hazard quotient (*mHQ*) (MacDonald et al., 2004) was calculated based on the following formula:

$$mHQ = \sqrt{\frac{C_i}{TEL_i} + \frac{C_i}{PEL_i} + \frac{C_i}{SEL_i}}$$

Where C_i – concentration of toxicant *i*, TEL_i – TEL value for toxicant *i*, PEL_i – PEL value for toxicant *i*, SEL_i – SEL value for toxicant *i*,

Toxic risk index (*TRI*) (Li et al., 2015) was calculated using the formula below:

$$TRI_i = \sqrt{\frac{\left(\frac{C_i}{TEL_i}\right)^2 + \left(\frac{C_i}{PEL_i}\right)^2}{2}}; TRI = \sum_{i=1}^{n} TRI_i$$

Where C_i – concentration of toxicant *i*, n – total quantity of analyzed toxicants, TEL_i – TEL value for toxicant *i*, PEL_i – PEL value for toxicant *i*.

Statistical analysis of data

Heavy metals and As in sediments were analyzed in three replicates. The results of the analyses were processed using Excel and Statistica Advanced 10 software packages: arithmetic mean, standard deviation were determined. Pollution and environmental risk indices were calculated based on average values.

RESULTS & DISCUSSION

Characteristics of sediments and average concentration of analyzed pollutants in bottom sediments of the coastal areas of Vostok Bay in 2015 and 2020 are presented in Table 2. Most sediments are sand (between 0.5 and 2 mm), which occupies a large part of the study area

Table 2. Fractional composition (%), concentration of total organic compounds (TOC, %), Heavy Metals and Arsenic in marine bottom sediments (µg/g)

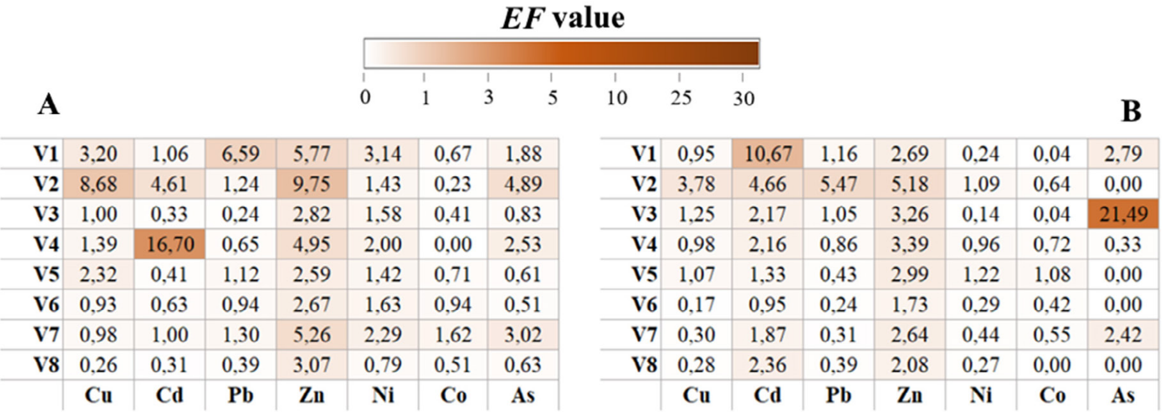
Years	Station	Fractional proportion grain size, %			TOC, %	Concentrations of pollutants, µg/g						
		Gravel	Sand	Silt and clay		Cu	Cd	Pb	Zn	Ni	Co	As
2015	V1	14.72	84.45	0.83	0.47	24.03±0.56	0.10±0.01	**123.73**±5.39	**162.58**± 10.31	29.50±1.01	5.00±0.23	3.50±0.20
	V2	9.58	78.72	11.71	1	**37.64±0.88**	0.25±0.03	13.40±0.58	**158.46**±10.05	7.74±0.27	1.00±0.05	5.24±0.30
	V3	13.33	82.27	4.39	1.72	6.04±0.14	0.02±0.003	3.57±0.16	64.07±4.06	11.99±0.41	2.50±0.12	1.25±0.07
	V4	82.21	16.94	0.85	2.16	3.32±0.08	0.50±0.06	3.90±0.17	44.43±2.82	6.00±0.21	bdl*	1.50±0.09
	V5	0	36.82	63.18	2.23	11.43±0.27	0.02±0.003	13.72±0.6	47.70±3.03	8.73±0.30	3.49±0.16	0.75±0.04
	V6	1.48	69.18	29.34	0.19	2.97±0.07	0.02±0.003	7.52±0.33	31.98±2.03	6.50±0.22	3.00±0.14	0.40±0.02
	V7	0.17	98.55	1.28	0.13	1.97±0.05	0.02±0.003	6.51±0.28	39.52±2.51	5.74±0.2	3.24±0.15	1.50±0.09
	V8	0	98.88	1.12	0.18	1.67±0.04	0.02±0.003	6.19±0.27	73.26±4.65	6.24±0.21	3.25±0.15	1.00±0.06
2020	V1	11.27	87.51	1.22	0.64	1.94±0.05	0.27±0.03	5.94±0.26	20.61±1.31	0.62±0.02	0.07±0.003	1.41±0.08
	V2	8.56	88.23	3.21	0.6	16.17±0.38	0.25±0.03	58.62±2.55	83.14±5.27	5.81±0.2	2.74±0.13	bdl
	V3	12.6	85.39	2.01	0.61	3.37±0.08	0.07±0.01	7.05±0.31	32.90±2.09	0.46±0.02	0.10±0.005	14.32±0.83
	V4	75.02	24.42	0.56	2.09	2.67±0.06	0.07±0.01	5.92±0.26	34.86±2.21	3.31±0.11	1.97±0.09	0.22±0.01
	V5	0	25.84	74.16	1.44	3.08±0.07	0.05±0.01	3.11±0.14	32.41±2.06	4.41±0.15	3.11±0.14	bdl
	V6	0.87	71.56	27.57	0.52	0.63±0.01	0.04±0.005	2.22±0.10	24.26±1.54	1.35±0.05	1.55±0.07	bdl
	V7	0.18	96.48	3.34	0.2	0.91±0.02	0.07±0.01	2.36±0.10	30.02±1.90	1.67±0.06	1.65±0.08	1.82±0.11
	V8	0.13	98.57	1.3	0.13	0.71±0.02	0.07±0.01	2.48±0.11	19.70±1.25	0.84±0.03	bdl	bdl
BC[1]		–	–	–	–	10	0.1	10	30	10	8	1.98
PC[2]		–	–	–	–	35	0.8	85	140	35	20	29
TEL[3]		–	–	–	–	18.70	0.68	30	124	15.9	–	7.24
PEL[3]		–	–	–	–	108	4.21	112	271	42.8	–	41.6
SEL[3]		–	–	–	–	110	10	250	820	75	–	33

*Below detection limit; [1]background concentrations; [2]permissible concentrations; [3]threshold effect level, probable effect level, severe effect le (between 66 and 99%)**.** The exceptions are station st. V4, where more than 70 % of the total mass was composed of particles larger than 2 mm (gravel) and st. V5 with a predominance of silt and clay (particles with a size less than 0.1 mm) fractions (more than 60%). The highest percentage of organic carbon (TOC) in both 2015 and 2020 was observed in the bottom sediments of the Srednyaya Cove (st. V3 and V4) and the mouth of the Volchanka River (st. V5).

The comparison of the obtained concentrations of heavy metals and As with background concentrations for Peter the Great Bay (Table 2) showed the following results. Exceeding background values of Zn content were observed at all stations in 2015 and at most stations in 2020. In samples from Gaydamak cove taken in 2015 exceeded the background for Cu, Cd, Pb, Ni and As, and in 2020 for Cu, Cd, Pb. In bottom sediments of the Srednyaya cove in 2015 Ni concentration was higher than background values, in 2020 the same picture was observed for As (7 times higher than background). Exceedance of permissible concentrations (PC) was recorded only in 2015 in the area of Gaydamak cove for Cu, Pb and Zn. The detected Cu, Zn, and Ni concentrations in the area were above the TEL threshold and the PEL for Pb. In 2020, Pb concentrations exceeding TEL values were recorded in Gaydamak cove and As – Srednyaya cove.

Analysis of the mean EF enrichment factor values (Fig. 2) of sediment samples across the Bay as a whole showed the following pattern in 2015: Zn (4.61) > Cd (3.13) > Cu (2.35) > As (1.86) > Ni (1.79) > Pb (1.56) > Co (0.64). The 2020 samples showed changes in the trends of heavy metal and As inputs to the Bay as shown by the following sequence: As (3.38) > Cd (3.27) > Zn (3.00) > Pb (1.24) > Cu (1.10) > Ni (0.58) > Co (0.43). The highest degree of enrichment of Cu, Pb, Zn, Ni and As in 2015 was observed in the area of Gaydamak cove, Cd - in Srednyaya cove, and Co in the mouth of the Litovka River. The EF value for Cd at st. V4 indicated a severe enrichment of this toxicant to the water area of Srednyaya cove. In sediment samples collected in 2020, the highest enrichment of Cu, Cd, Pb, and Zn was recorded in the Gaydamak cove, As – in Srednyaya cove, Ni and Co – in apex of Bay. For Cd at st. V1 and As at st. V3 was noted severe enrichment.

The results of the modified hazard quotient (*mHQ*) calculation (Fig. 3) for the 2015 sediment samples showed negligible to low levels of potential toxic effects of the analyzed pollutants. However, lead content in sediments of Gaydamak cove (st. V1) conformed to the 5th class (considerable severity of contamination), and Ni - to the 4th class of hazard (moderate severity of contamination). A similar pattern was observed in the 2020 samples, characterized by low toxicant hazards. Concentrations of lead and arsenic in sediments from Gaydamak and Srednyaya coves, based on the *mHQ* index calculation, were at the level of hazard class 4, which corresponded to moderate severity of contamination.



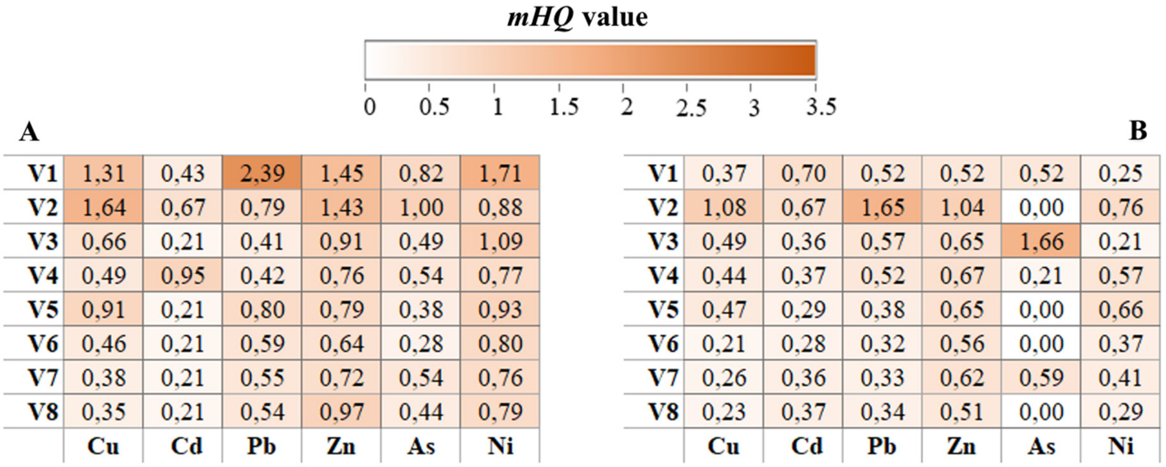
A

	Cu	Cd	Pb	Zn	Ni	Co	As
V1	3,20	1,06	6,59	5,77	3,14	0,67	1,88
V2	8,68	4,61	1,24	9,75	1,43	0,23	4,89
V3	1,00	0,33	0,24	2,82	1,58	0,41	0,83
V4	1,39	16,70	0,65	4,95	2,00	0,00	2,53
V5	2,32	0,41	1,12	2,59	1,42	0,71	0,61
V6	0,93	0,63	0,94	2,67	1,63	0,94	0,51
V7	0,98	1,00	1,30	5,26	2,29	1,62	3,02
V8	0,26	0,31	0,39	3,07	0,79	0,51	0,63

B

	Cu	Cd	Pb	Zn	Ni	Co	As
V1	0,95	10,67	1,16	2,69	0,24	0,04	2,79
V2	3,78	4,66	5,47	5,18	1,09	0,64	0,00
V3	1,25	2,17	1,05	3,26	0,14	0,04	21,49
V4	0,98	2,16	0,86	3,39	0,96	0,72	0,33
V5	1,07	1,33	0,43	2,99	1,22	1,08	0,00
V6	0,17	0,95	0,24	1,73	0,29	0,42	0,00
V7	0,30	1,87	0,31	2,64	0,44	0,55	2,42
V8	0,28	2,36	0,39	2,08	0,27	0,00	0,00

Fig.2. Enrichment factor (*EF*) values for sediment samples collected in 2015 (A) and 2020 (B) in the Vostok Bay



A

	Cu	Cd	Pb	Zn	As	Ni
V1	1,31	0,43	2,39	1,45	0,82	1,71
V2	1,64	0,67	0,79	1,43	1,00	0,88
V3	0,66	0,21	0,41	0,91	0,49	1,09
V4	0,49	0,95	0,42	0,76	0,54	0,77
V5	0,91	0,21	0,80	0,79	0,38	0,93
V6	0,46	0,21	0,59	0,64	0,28	0,80
V7	0,38	0,21	0,55	0,72	0,54	0,76
V8	0,35	0,21	0,54	0,97	0,44	0,79

B

	Cu	Cd	Pb	Zn	As	Ni
V1	0,37	0,70	0,52	0,52	0,52	0,25
V2	1,08	0,67	1,65	1,04	0,00	0,76
V3	0,49	0,36	0,57	0,65	1,66	0,21
V4	0,44	0,37	0,52	0,67	0,21	0,57
V5	0,47	0,29	0,38	0,65	0,00	0,66
V6	0,21	0,28	0,32	0,56	0,00	0,37
V7	0,26	0,36	0,33	0,62	0,59	0,41
V8	0,23	0,37	0,34	0,51	0,00	0,29

Fig. 3. Modified hazard quotient (*mHQ*) values for sediment samples collected in 2015 (A) and 2020 (B) in the Vostok Bay

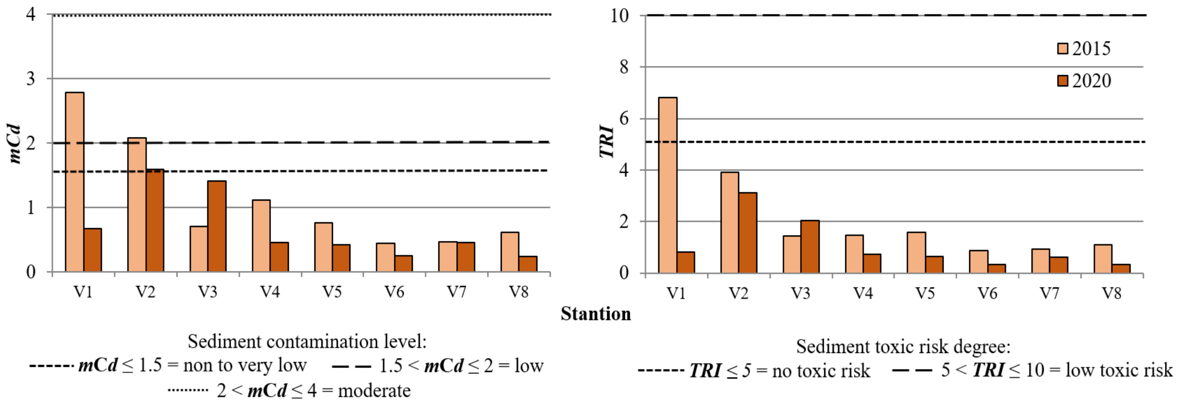


Fig. 4. Modified Contamination Degree Index (*mCd*) and Toxic Risk Index (*TRI*) values for sediment samples collected in 2015 and 2020 in the Vostok Bay

Calculation of modified contamination degree (*mCd*) and toxic risk (*TRI*) indices of samples (Fig.4) collected in 2015 showed moderate level of contamination and low toxicity of sediments in Gaydamak cove. The sediments collected in 2020 were characterized by very low degree of contamination and no toxic effect.

It is known that active accumulation of organic carbon and biogenic elements in bottom sediments is associated with a common source of input – remains of plants and planktonic organisms (Sevastyanov et al., 2020). In the summer period of 2015 in waters of Vostok Bay the increased BOD_5 value was observed in the estuarine zones of the Volchanka and Litovka rivers (2.17 – 3.22 mg/l). In the waters of Srednyaya and Vostok coves, the concentration of phosphates, with predominance of the organic form, reached up to 200 µg/L (Grigorieva et al., 2020). The maximum values of organic matter (OM) in the bottom layer of Vostok Bay in 2020 (by BOD_5 values) were observed in the Srednyaya Bay (1.97 mg/l) and the mouth of the Volchanka River (1.68 mg/l). These areas also had higher levels of chlorophyll *a* (2.7 and 2.4 µg/dm^3, respectively) in bottom water layers (Khristoforova et al., 2023). This may indicate the contribution of phytoplankton to the total organic matter content of bottom sediments. The ratio of organic to inorganic phosphorus (P_{org} : P_{min}) in the bottom water layer of these areas was characterized by the predominance of the mineral form, which confirms the degradation of OM. The maximum P_{org} content for these areas was 25 μg/L. The levels of TOC in bottom sediments of Srednyaya and Vostok coves confirm the active input of organic matter into the waters of these areas.

The entry of OM into the surface layer of bottom sediments occurs in the process of realization of one of three main mechanisms: deposition of organic suspended particles and further co-burial with inorganic forms of suspended matter; sedimentation of suspended OM and its burial as a result of the process of biological mixing (bioturbation); accumulation of OM by benthic organisms with further burial in deeper sediment layers in the form of life products or dead OM (Tishchenko et al., 2020). The main source of organic matter input in the area of station V5 is the runoff of the Volchanka River, and the predominance of silty fractions in the sediment structure contributes to active accumulation (Table 1). The maximum content of heterotrophs in the summer period of 2020 equal to 105 CFU/mL was observed in the estuary zone of the Volchanka River, which is directly related to the inflow of domestic wastewater. In this area, as well as in the apex part of the Srednyaya cove settlements of *Zostera japonica* (Ascherson & Graebner, 1907) and *Zostera asiatica* (Miki, 1932) are located at a depth of 0.5 to 1.2 m. They are replaced with extensive thickets of *Zostera marina* (Linnaeus, 1753) with biomass reaching 2150 g/m2 (average biomass - 1162.9 g/m2) (Galysheva, 2004; Kozhenkova, 2008). Suspended matter produced by thickets is a major source of OM inputs to the bottom sediments of Tikhaya Zavod' and Srednyaya coves (Tishchenko et al., 2020).

Levels of organic matter in sediments can indicate the status of benthic flora and fauna. Decrease and reorganization of qualitative and quantitative characteristics of communities are observed at TOC value more than 2.5 %. Under these conditions, elimination of K-strategist species is noted with gradual replacement by more tolerant forms of R-strategists. In such sediments they note the occurrence of sulfate-reduction processes and reduction of oxygen content, which leads to the production of hydrogen sulfide, which is harmful to most hydrobionts (Ovsyanyi et al., 2009). The levels of TOC detected in sediments of Srednyaya cove in 2015 and 2020 are close to the critical threshold content and may indicate the development of adverse effects on biota.

Comparison of our obtained concentrations of metals in sediments sampled in 2015 with the results of sampling in the late 1990s (Khristoforova et al., 2004) showed a decrease in Cu, Pb, Zn, and Ni in the surface sediment layer of the northern part of the bay (Srednyaya and Vostok coves). The content of Cu, Zn and Ni in the bottom sediments of Gaydamak cove was higher or remained at the same level. Compared to 2015, the levels of modern concentrations (for 2020) of most of the pollutants we analyzed in the Bay sediments are an order of magnitude lower. The analysis of heavy metal concentrations detected in different marine coastal areas of the Pacific Asia region (Table 3) showed that the content of these elements in the sediments of Vostok Bay is not high.

The main contributors of pollutants to the waters of Vostok Bay are the port and industrial areas adjacent to the ship repair and fish processing enterprises in Gaydamak cove, as well as extensive recreational areas located along the entire coast of the Bay. The Volchanka River makes a significant contribution to pollution of these area. The impact of river flow increases in the spring and summer period, which is predominantly associated with flooding periods (Grigorieva et al., 2020). The high concentrations of Cd, Zn, Cu and As detected as a result of our study in Gaydamak and Srednyaya coves may be related to diesel and fuel oil combustion, as a result of active exploitation of the marine fleet and power generators, and Pb - as a result of ship repair enterprises. The results of microbial indication carried out in summer 2020 (Khristoforova et al., 2023) showed high abundance of metal-resistant microorganisms in the waters of Gaydamak cove indicating water pollution by Cu, Zn and Pb The maximum number of Cu, Ni and Zn - resistant bacteria was observed in the area of the Volchanetskaya channel

Table 3. Heavy metals and arsenic concentrations (μg/g) in bottom sediments from different marine coastal areas of the Pacific Asia region

Location	Cu	Cd	Pb	Zn	Ni	Co	As	Reference
Sea of Japan, Vostok Bay, Russia (2015/2020)	1.67-37.64/ 0.63-16.17	0.02-0.5/ 0.04-0.27	3.57-123.73/ 2.22-58.62	31.98-162.58/ 19.7-83.14	5.74-29.5/ 0.46-5.81	1-5/ 0.07-3.11	0.4-5.24/ 0.22-14.32	This study
Sea of Japan, Vostok Bay, Russia	14-27	–	45-125	95-359	19-46	–	–	Khristoforova et al., 2004
Sea of Japan, open areas of Amur and Ussuri Bays, Russia	1.2-9.5	0.1-1.4	2.8-13	9.3-41	3.9-14	0-4.4	–	Moshchenko et al., 2019
Sea of Japan, inland bay areas of Amur and Ussuri Bays, Russia	9-19	0.1-2.2	6.8-25.5	51-91	8.5-33	1.8-11	–	
Sea of Japan, coastal zones of Vladivostok, Russia	24-70	0.1-2.9	28-54	63-139	8.3-31	1.6-70.5	–	
Sea of Japan, Golden Horn and Diomid Bays, Russia	249-319	3.1-7.8	154-210	381-471	20-20	3.1-5.6	–	
Bay of Bengal, Bay of Bengal, East Coast of India	1.25- 45.59	1.85- 8.39	1.15- 11.83	12.22- 43.82	2.79- 42.33	–	–	Lakshmanna et al., 2023
South China Sea, Northwest coast of Sabah, Malaysia	0.41- 40.70	–	0.28- 9.56	3.50- 51.38	0.65- 225.32	0.08- 18.03	–	Ling et al., 2023
North Yellow Sea, Weihai Bay, China	3.52–44.60	0.041–0.42	17.9–41.3	7.59–202.0	–	–	–	Shang et al., 2023
South China Sea, Hsingda Harbor, Southwestern Taiwan	6.6- 77.7	–	10.6-50.4	62.6-309	19.8-34.0	–	–	Lim et al., 2023
Inner shelf of East China Sea, east China	5.77-40.8	35-96.4	17.3-53.7	45.6-143	24.9-55.6	–	3.8-51.1	Wang et al., 2021
Yellow Sea, Bohai Bay, China	3.3-45.6	0-0.5	9.7-33.9	16.7-130.4	–	–	3.3-33.6	Ding et al., 2019

and the mouth of the Volchanka River. These facts indicate an active input of heavy metals not only from the industrial area of the western coast of the bay, but also from its apex part due to river and terrigenous runoff. Copper and zinc are essential elements for living organisms and are characterized by high biological absorption coefficients. The planktonic group of organisms is characterized by active accumulation of cadmium and lead, typical pollutants of the marine environment (Khristoforova et al, 2023).

Marine currents play a major role in the transport and accumulation of pollutants. Surface and near-bottom currents directed from the apex part of the bay along the western coast (Gaiko, 2017) contribute to the migration of pollutants coming with the river flow of the Volchanetskaya channel, Volchanka River and large industrial cluster located on the western coast of bay. Local cyclonic circulation in the coves of Srednyaya and Gaydamak, caused by the indented coastline, influences the processes of burial and accumulation of pollutants by bottom sediments. It should be noted that the general pattern of water mass movement in Vostok Bay is not stable and can change both in time and in area. This depends on many factors such as wind, tides, waves, atmospheric pressure, river flow, solar radiation, and typhoons (Gaiko, 2017). For example, in the spring-summer period, the influence of the summer monsoon with the dominance of southeastern wind is noted. This fact together with the increase in the Volchanka River flow leads to the effect of river water locking by wind surge in the north-western part of the bay in the area of Srednyaya cove (Khristoforova et al, 2023).

The obtained individual values of modified hazard quotient (*mHQ*) for Pb, Cu, Ni and As may indicate a high potential toxic effect of concentrations of these elements for benthic fauna inhabiting the sediments of Gaydamak and Srednyaya coves. The ability of heavy metals to form stable complexes with amino acids, as well as other molecules containing thio-(SH-) or alkylthiogroupings (RS-), leads to inhibition of enzyme activity, weakening of the body's defense functions and development of pathological conditions in hydrobionts (Kantserova et al., 2016).

Calculations of complex indices (*mCd* and *TRI*) for bottom sediments of Gaydamak cove sampled in 2015 showed that despite exceeding the permissible concentrations of Cu, Pb and Zn and high *mHQ* indices, the overall pollution index was at an average level, with low potential toxicity to hydrobionts. The values of integral indices for 2020 samples showed a decrease in pollution and toxicity indices for most stations, except for station 3, in the area of Srednyaya cove. In this area there was an increase in index values compared to 2015, associated with high arsenic content in the studied sediments.

CONCLUSION

The results of the study demonstrated a general decrease in the level of contamination of bottom sediments with heavy metals and arsenic and their potential toxicity to hydrobionts. The exception is cadmium, which increased, albeit insignificantly, in the bottom sediments of most of the study areas. The continuous process of cadmium input to the marine coastal areas of the Vostok Bay may be related to the combustion of diesel, heavy motor and boiler fuels, as well as fuel oil due to the operation of commercial and passenger vessels, industrial furnace and installations, heating systems.

The western coast of the Bay (Gaydamak and Srednyaya coves) remains the most polluted area, which correlates with the results of previous years studies (Khristoforova et al., 2004). The highest concentrations of Cu, Pb, Zn, Ni, Co and As were found in bottom sediment samples collected from Gaydamak Cove in 2015. We assume that the exceedance of the permissible concentrations of Pb and Zn at station V1 is linked to the sudden introduction of these pollutants as a result of the modernisation of the fishing and fish processing facilities, which has been linked to the rebranding of the company since 2014. Same situation may be the cause of the high concentrations of Cu and Zn in the vicinity of station V2. Since 2014, after a break of several years, the ship repair facilitie, located on the coast near the V2 station, has been carrying out projects not only for the maintenance and repair of small and medium-sized vessels, but also for the construction of cargo-passenger barges and ferries. It is important to note that heavy metal accumulation in this area may be favoured by the highly indented coastline of the cove, which results in poor water exchange with the waters of Vostok Bay. The 2020 sampling results showed a decrease in heavy metals in the bottom sediments of the area. This may indicate that, despite the increase in industrial production by companies, the implementation of national and regional environmental legislation and the necessary treatment of industrial waste water are helping to limit heavy metals in the cove.

Comparison of the concentrations of pollutants with those for different marine coastal areas of the Pacific Asia region showed that the Vostok Bay has low levels concentrations of heavy metals and As. This may be due to the fact that although ship repair and fish processing facilities are active on the west coast of the Bay, they are relatively small in terms of the total services provided compared to international ports and extractive industries, and serve only the local needs of the country's domestic market.

Vostok Bay is becoming a more attractive place every year for both industrial and recreational activities. As a result of activity of fishing and ship repair industries, ports, as well as recreation centers, the water area receives a large amount of pollutants of both inorganic and and organic nature. The building of a mariculture base in the southwestern part of the Bay (Khristoforova et al., 2023), growth in industrial output of ship repair and fish processing enterprises, as well as the possible resumption of works on the construction of facilities of Rosneft's Eastern Petrochemical Complex (EPCC) (On the Preparation of Documentation…, 2020) in its southeastern part will inevitably lead to even greater inputs of pollutants into the marine environment. High local variability of hydrologic and hydrochemical parameters may contribute to active accumulation of pollutants in the bottom sediments of the coves. Therefore, it is important to continue further monitoring studies to control heavy metals and arsenic in sediments.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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