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ASSESSMENT OF HEAVY METAL POLLUTION OF COASTAL WATERS OFF THE MURAVYOV-AMURSKY PENINSULA USING ALGAE AS BIOINDICATORS

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Fe, Mn, Cu, Zn, Pb, Cd, and Ni concentrations were measured in brown algae [*Sargassum miyabei* Yendo and *S. pallidum* (Turner) C. Agardh] and green algae [*Blidingia minima* (Nägeli ex Kützing) Kylin, *Ulva lactuca* Linnaeus, and *U. linza* Linnaeus] sampled in July 2017 in coastal waters off the city of Vladivostok, Muravyov-Amursky Peninsula, Sea of Japan. Heavy metal concentrations in algae were determined by atomic absorption spectroscopy after thalli mineralization with nitric acid. Dissolved trace elements in seawater were measured by ultrafiltration of water samples and $CHCl_3$–DDTK-Na method. The degree of pollution in various areas of the coastal zone was assessed applying the hazard coefficient for algae (K_H). It was calculated as the ratio of metal concentration in an alga to the upper threshold level of background concentrations of the element. Also, integral Trace Element Pollution Index (TEPI-threshold) was applied using $K_H \geq 1$. Coastal waters off Vladivostok were slightly polluted by heavy metals. At stations located north and south from a solid waste landfill, TEPI-threshold was 2.4–2.8 due to pollution by Pb and Cu (2.7–12 $C_{threshold}$), as well as Zn, Fe, Mn, and Ni. Algae from upper areas of the Amur and Ussuri bays were Fe- and Mn-enriched because of river discharge; TEPI-threshold was 1.7–3.0. Macrophytes of the Eastern Bosphorus Strait were polluted by Fe (3–10 $C_{threshold}$), as well as Mn, Cu, Zn, and Ni (1–1.5 $C_{threshold}$), which results from port activities, shipping, and construction of bridges; TEPI-threshold was 1.0–2.1. Off the eastern coast of the Muravyov-Amursky Peninsula, there was a local zone of high-degree pollution formed due to rainwater drainage from the reclaimed solid waste landfill in Vladivostok; TEPI-threshold was 16. Out of heavy metals studied, Fe and Cu were main pollutants at this station ($K_H > 80$ in algae), while Pb, Mn, Zn, and Ni were co-pollutants. In seawater at this station, concentrations of dissolved elements exceeded the background levels, and pollution by Cu was equal to 3 MPC for fishery reservoirs.

Keywords: heavy metals, brown algae, green algae, Amur Bay, Ussuri Bay, Peter the Great Bay, Muravyov-Amursky Peninsula, Sea of Japan

Common species of marine algae have long been successfully used as bioindicators of metal and nonmetal pollution in the marine environment [Aboal et al., 2023; Bryan, Hummerstone, 1973; Malea, Kevrekidis, 2014; Obluchinskaya et al., 2013; Pan et al., 2018; *etc.*]. Their application is based on the relationship between metal content in the environment and organisms [Rainbow, 2020; Rainbow, Phillips, 1993; *etc.*].

Periodic monitoring of heavy metal concentrations in brown algae from the northwestern Sea of Japan has been carried out since 1976 [Khristoforova, 1989]. Spatial and interannual assessments of metal pollution in coastal waters based on algal data have been obtained for the open sea coast [Shul'kin et al., 2015], Peter the Great Bay, its pristine spots, and areas under significant anthropogenic load [Chernova, Kozhenkova, 2016; Kozhenkova et al., 2006, 2021].

Several localized pollution sites exist in coastal waters of the Russian part of the Sea of Japan [Kozhenkova et al., 2021; Shulkin, 2004]. One of them covers the northwestern Peter the Great Bay off the Muravyov-Amursky Peninsula which is home to the Vladivostok agglomeration.

The aim of this study was to assess heavy metal pollution in coastal waters off the Muravyov-Amursky Peninsula by analyzing content of trace elements in brown algae [*Sargassum miyabei* Yendo, 1907 and *S. pallidum* (Turner) C. Agardh, 1820] and green algae [*Blidingia minima* (Nägeli ex Kützing) Kylin, 1947, *Ulva lactuca* Linnaeus, 1753 (= *U. fenestrata* Postels et Ruprecht), and *U. linza* Linnaeus, 1753].

MATERIAL AND METHODS

Brown algae *S. miyabei* and *S. pallidum* and green algae *U. lactuca*, *U. linza*, and *B. minima* were sampled from coastal waters off the Muravyov-Amursky Peninsula: in the western Amur Bay, the eastern Ussuri Bay, and the Eastern Bosphorus Strait. A total of 27 stations were surveyed (Fig. 1). *S. miyabei* was sampled at 16 stations; *S. pallidum*, at 16 stations; *U. linza*, at 8 stations; *U. lactuca*, at 3 stations; and *B. minima*, at 3 stations.

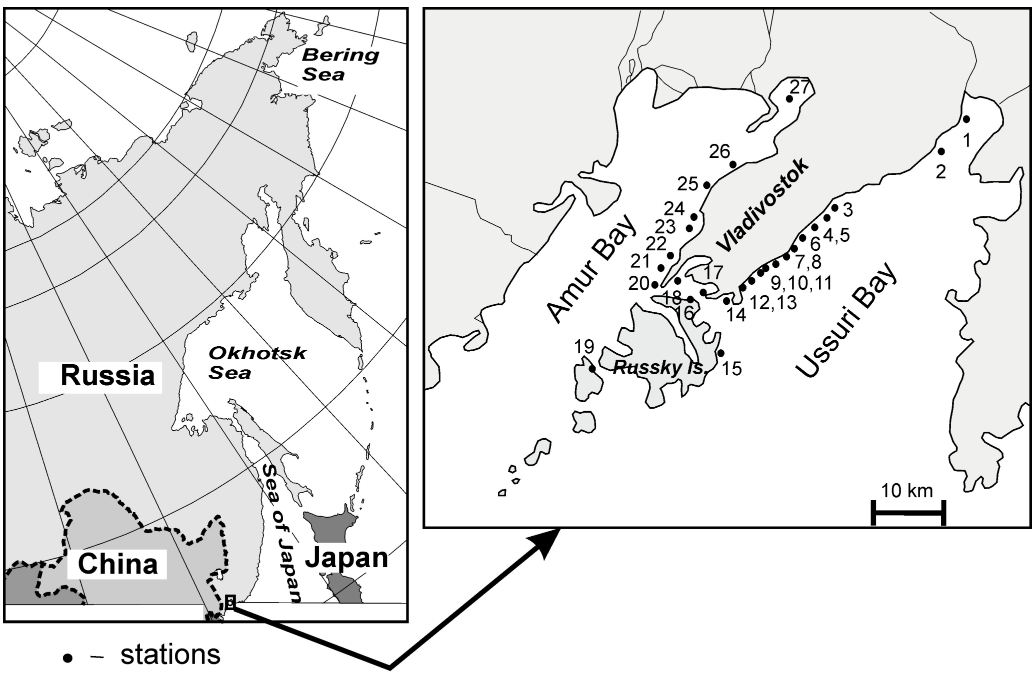

Fig. 1. Sampling stations off the Muravyov-Amursky Peninsula. The Ussuri Bay: 1, Cherepakha Cape; 2, Muravyiny Cape; 3, Lazurnaya Cove, Zeleny Cape; 4, "Politekhnik" recreational base; 5, "Zhemchuzhina" recreational base; 6, Desantnaya Cove; 7, "Gornostay" solid waste landfill; 8, Rybachy village; 9, Gornostay Cove; 10, Promezhutochnaya Cove, north; 11, Promezhutochnaya Cove, south; 12, Sukhoputnaya Cove; 13, Sobol Cove; 14, Patrokl Cove. The Russky Island: 15, Akhlestyshev Cape; 16, Pospelov Cape. The Eastern Bosphorus Strait: 17, Nazimov Cape; 18, Tokarevskaya Koshka Cove. The Popov Island: 19, Stark Strait. The Amur Bay: 20, Tokarevsky Cape; 21, Primorskaya Railway harbor; 22, Bezymyannaya Cove; 23, Kirpichny Zavod Cove; 24, a site between Firsov Cape and Grozny Cape; 25, Krasny Cape; 26, Sanatornaya railway station; 27, Uglovoy Cove

Algae were sampled on 10–17 and 25–27 July, 2017, at depths of 0.5–1.5 m. Depending on size, 5–15 specimens of each species were sampled, washed with seawater, and packed in polyethylene bags. In a laboratory, we cleaned algae of epiphytes and invertebrates, formed 5 samples, and dried them at +85 °C. Dried samples were ground, and 0.5-g subsamples were mineralized in a microwave system with 6 mL of high-purity nitric acid. Concentrations of Fe, Mn, Cu, Zn, Pb, Cd, and Ni were determined by flame and flameless (Pb) atomic absorption spectrometry on a Shimadzu AA-6800 spectrometer at the core facility "Center for Landscape Ecodiagnostics and GIS Technologies" (Pacific Geographical Institute FEB RAS). Quality control for sample preparation and trace element determination was performed using blank samples and international certified reference material (Table 1). Concentrations are presented in $\mu g \cdot g^{-1}$ dry weight.

Table 1. Analysis of standard reference material "Leaf of Birch" (LB-1, GSO 8923-2007, Irkutsk) and "Oyster" (NBS Oyster 1566a)

Parameter	Metal content, $\mu g \cdot g^{-1}$						
	Cu	Mn	Fe	Zn	Cd	Pb	Ni
"Leaf of Birch"							
Certified reference sample	7.3 ± 0.6	930 ± 70	730 ± 70	94 ± 6	0.16 ± 0.03	3.7 ± 0.6	5.8 ± 0.8
Result of control determination	7.4	835	765	95.2	0.13	3.15	6.3
"Oyster"							
Certified reference sample	66.3 ± 4.3	12.3 ± 1.5	539 ± 15	830 ± 57	4.15 ± 0.38	0.37 ± 0.014	2.25 ± 0.44
Result of control determination	64	11.4	517	903	4.65	0.34	2.09

Seawater was sampled in plastic canisters from the undersurface layer at 13 stations. On the same day, 1 L of each sample was filtered through a 0.45-µm membrane filter in the laboratory. Metal complexes were concentrated from 1 L of filtrate using a chloroform–DDTK-Na system. Suspended matter was determined by weighing filters before filtration and after it. Concentrations of dissolved metals were established by atomic absorption spectrometry on a Shimadzu AA-6800 spectrophotometer. Quality of determining Pb and Cd content in water and Pb in algae was additionally controlled using standard additions, with recoveries ranging 80–85%. Salinity was measured conductometrically.

For each station, arithmetic mean and standard deviation of concentrations of trace elements were calculated for algal samples consisting of 5 specimens.

Heavy metal contamination in the Amur and Ussuri bays was assessed by comparing measured concentrations in algae from this study with published upper threshold values of background ranges for corresponding species: $C_{threshold}$ (Table 2). These threshold values were derived as the median plus double median absolute deviation from the sample median (median + 2 MAD) [Chernova, 2012; Chernova, Kozhenkova, 2016] and subsequently validated [Chernova, Shulkin, 2019].

For *U. linza* and *B. minima*, threshold values for metals were adopted from those for *U. lactuca* [Kozhenkova, Chernova, 2017]. The hazard coefficient (K_H) for pollution of an area by an element, defined as the ratio of the *i*-th metal concentration in an alga (C_i) to $C_{threshold}$, was calculated: $K_H = C_i / C_{threshold}$. Stations with $K_H > 1$ were considered polluted.

Table 2. Median and threshold metal concentrations ($C_{threshold}$) in brown algae (*Sargassum miyabei* and *Sargassum pallidum*) and green alga (*Ulva lactuca*) of coastal waters of the Sea of Japan (μg·g^{-1} of dry mass)

Parameter	Taxon	Cu	Mn	Fe	Zn	Pb	Cd	Ni
Median* / $C_{threshold}$	*S. miyabei*	2.9 / 4.7	266 / 714	353 / 746	17.0 / 23.9	0.8 / 1.8	1.6 / 2.9	2.3 / 3.6
	S. pallidum	2.3 / 3.9	168 / 455	317 / 672	15.0 / 23.8	0.6 / 1.5	1.1 / 1.7	2.0 / 3.8
	U. lactuca	4.1 / 7.3	17.2 / 34.6	317 / 672	7.6 / 13.9	1.3 / 3.0	0.07 / 0.15	1.6 / 3.5
World median** / Q3	brown algae	5.7 / 13.0	67 / 135	301 / 848	49 / 120	5.5 / 11.0	1.0 / 2.15	6.0 / 11.4
	green algae	7.1 / 12.6	81 / 182	492 / 1,270	36 / 60.6	5.2 / 11.6	0.42 / 0.90	3.9 / 7.1

Note: *, [Chernova, Kozhenkova, 2016; Kozhenkova, Chernova, 2017]; **, [Sánchez-Quiles et al., 2017]. Q3 is the element concentration corresponding to the value of the third quartile of the sample.

A comprehensive assessment was conducted using the Trace Elements Pollution Index, TEPI [Richir, Gobert, 2014] modified as follows:

$$\text{TEPI-threshold} = (Cf_1 \times Cf_2 \times \ldots \times Cf_n)^{1/n},$$

where Cf_1, Cf_2… Cf_n are normalized concentrations relative to $C_{threshold}$ (Cf_n / $C_{threshold}$);

n is the number of trace elements with $C_i \geq C_{threshold}$.

Stations with TEPI-threshold > 1 were considered polluted.

RESULTS

The hazard coefficients (K_H) for metal pollution in algae from waters off Vladivostok are presented in Figs 2 and 3.

Stations with metal concentrations in *Sargassum* spp. exceeding threshold levels were located in the upper part of the Amur Bay [sta. 26 and 27 (Mn, Fe, and Ni)], in coastal waters off the city [sta. 22 and 23 (Zn and Ni)], in the contact zone between the Amur and Zolotoy Rog bays [sta. 20 (Fe, Mn, and Zn)], and in the Eastern Bosphorus Strait [sta. 19 (Mn, Fe, and Pb)] (Fig. 2). In the Ussuri Bay, metal pollution was noted in algae growing near the "Gornostay" solid waste landfill reclaimed in 2011 [sta. 6–8 (Cu, Mn, Fe, Zn, Pb, Ni, and Cd), sta. 9 (Cu, Zn, Pb, and Ni), and sta. 10 (Ni)], as well as in the upper part of the bay [sta. 1 and 2 (Fe, Mn, and Ni)] (Fig. 3).

The highest K_H values for Fe and Cu were observed in a green alga *B. minima* at sta. 7: values exceeded $C_{threshold}$ by factors of 276 and 82, respectively (Figs 2, 3). Concentrations of Zn, Ni, Pb, Cd, and Mn corresponded to 29, 26, 13, 3, and 3 K_H. Ni content exceeded $C_{threshold}$ in algae at 50% of the stations.

Concentrations of dissolved metals in water from sampling sites were elevated relative to background levels for Peter the Great Bay only at sta. 7 (Mn, Cu, Zn, Cd, Pb, and Ni) and sta. 6 (Zn and Pb). Cu content in water at sta. 7 exceeded $MPC_{fishery}$ (maximum permissible concentration for fishery reservoirs) by a factor of 3 (Table 3). The amount of suspended matter was the highest at sta. 1, near the mouth of the Artemovka River.

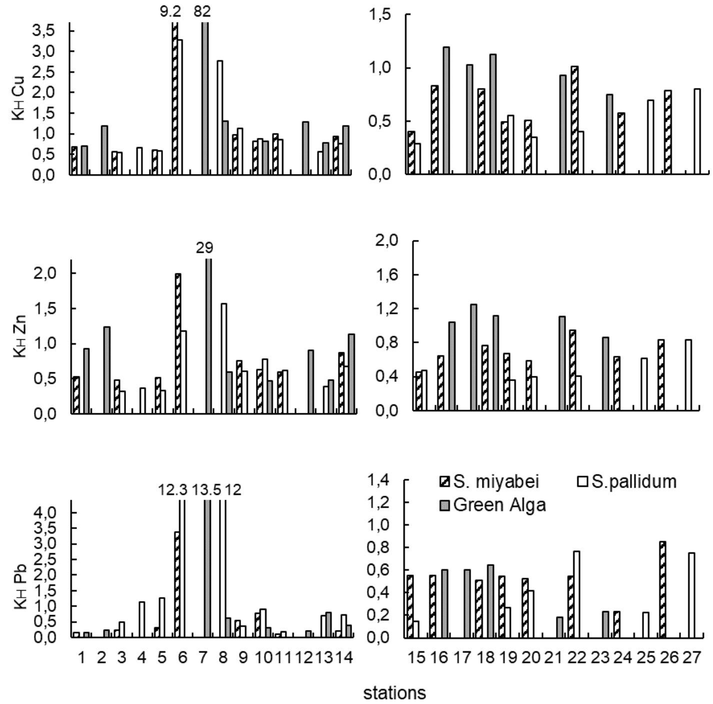

Fig. 2. Hazard coefficient of algae pollution by Cu, Zn, and Pb ($K_H = C_i / C_{threshold}$) (station numbers are as in Fig. 1)

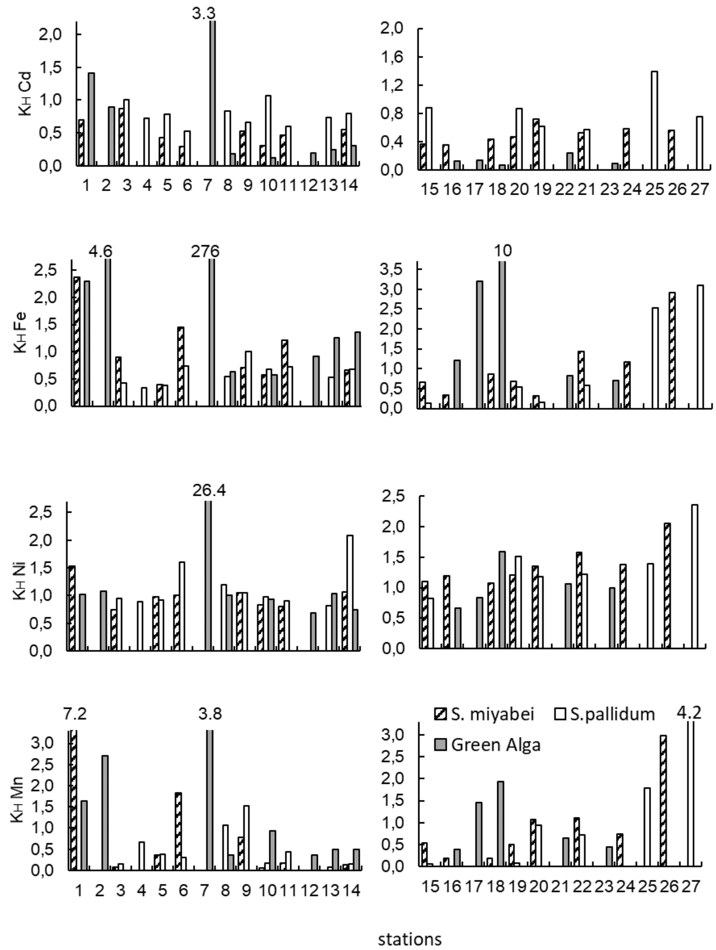

Fig. 3. Hazard coefficient of algae pollution by Cd, Fe, Ni, and Mn ($K_H = C_i / C_{threshold}$) (station numbers are as in Fig. 1)

Table 3. Concentrations of dissolved forms of metals ($\mu g \cdot L^{-1}$), salinity (S, ‰), and turbidity (D, $mg \cdot L^{-1}$) in the water environment of the Ussuri and Amur bays

Station	No.	Date	S	Fe	Mn	Cu	Zn	Cd	Ni	Pb	D
Cherepakha Cape	1	12.07.2017	16.59	1.44	2.12	0.51	1.46	0.009	0.62	0.025	8.68
Muravyiny Cape	2	12.07.2017	24.93	0.59	5.48	0.55	0.86	0.015	0.64	0.029	4.59
Desantnaya Cove, south	6	13.07.2017	32.09	2.08	3.81	0.47	2.73	0.041	0.91	0.238	2.95
Solid waste landfill	7	13.07.2017	31.49	1.24	44.62	16.92	8.62	0.123	1.42	0.159	3.36
Rybachy village	8	12.07.2017	32.32	0.77	1.01	1.36	1.50	0.022	0.64	0.031	3.00
Gornostay Cove	9	13.07.2017	30.62	0.38	3.96	0.66	0.95	0.019	0.57	0.015	4.45
Sukhoputnaya Cove	12	12.07.2017	32.15	0.95	0.89	0.91	0.89	0.020	0.56	0.020	3.14
Patrokl Cove	14	12.07.2017	32.47	1.53	0.64	0.59	2.27	0.021	0.47	0.008	3.04
Stark Strait	19	30.07.2017	28.46	2.92	0.84	1.03	0.49	0.009	0.80	0.019	2.06
Pospelov Cape	16	10.07.2017	30.68	1.52	1.95	0.75	1.04	0.017	0.60	0.017	0.95
Tokarevsky Cape	20	13.07.2017	31.10	1.34	0.77	0.42	0.63	0.012	0.64	0.021	1.09
Primorskaya Railway harbor	21	13.07.2017	30.26	0.89	2.23	0.48	0.90	0.014	0.68	0.022	0.82
A site between Firsov Cape and Grozny Cape	24	10.07.2017	29.07	1.81	6.45	0.41	1.05	0.015	0.63	0.021	1.73
$MPC_{fishery}$			–	100	50	5	50	1	10	6	–
Background for open/sheltered areas of Peter the Great Bay*			–	–	10	0.3 / 1.2	0.5 / 0.8	0.05 / 0.04	0.2 / 1.1	0.05 / 0.06	–

Note: *, [Shulkin, 2004; Shulkin et al., 2013]. A dash denotes no data.

TEPI-threshold values calculated considering only elements with concentrations equal to $C_{threshold}$ or exceeding it are presented in Fig. 4. TEPI-threshold values determined considering all seven studied elements are given in parentheses below.

The highest level of pollution in algae by seven metals, TEPI-threshold = 16, was recorded at sta. 7 (at the solid waste landfill) (Fig. 4). TEPI-threshold at sta. 6 and 8 (in the landfill vicinity) was reduced to 2.4–2.8 (1.6–1.7 when accounting for seven metals). In upper areas of the Ussuri and Amur bays, the index was 1.7–3.0 (1.0–1.2) and 2.6–3.0 (1.2–1.4), respectively. At all other stations off the Muravyov-Amursky Peninsula, TEPI-threshold remained within 1 (< 1 when considering concentrations of seven metals).

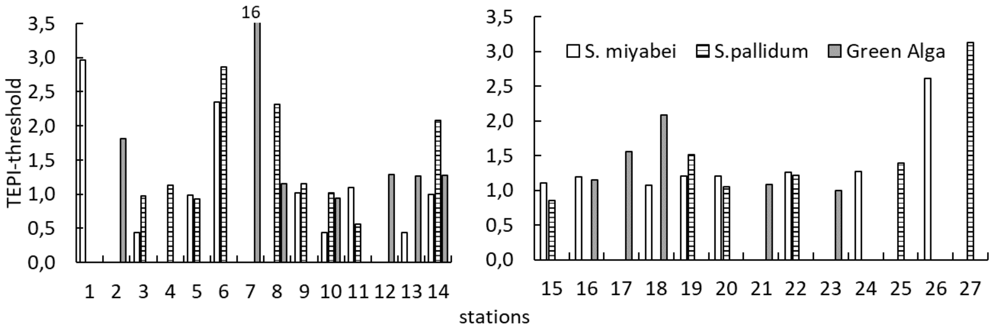

Fig. 4. Trace Element Pollution Index (TEPI-threshold) values for algae off the Muravyov-Amursky Peninsula (station numbers are as in Fig. 1)

DISCUSSION

Sources of metal input into the marine environment off Vladivostok include riverine discharge, atmospheric deposition, municipal and industrial wastewater, surface runoff from urbanized areas, leachate from solid waste landfills, ash ponds, and tailings storage facilities. Also, trace elements can enter the marine environment because of corrosion of port infrastructure facilities and ship hulls, sunken vessels, dredging operations, and sea dumping of dredged material [Kozhenkova et al., 2021; Shul'kin et al., 2017].

Previous studies of coastal waters off Vladivostok revealed that seawater is contaminated with petroleum hydrocarbons, phenols, and organic matter from domestic sewage. Oil pollution in open areas of the Amur and Ussuri bays occurs due to the discharge of ballast and bilge waters from vessels stemming from the absence or insufficient capacity of onshore oil treatment facilities [Marine Water Pollution, 2018]. Bottom sediments off the city are contaminated with petroleum hydrocarbons, phenols, pesticides, cadmium, and mercury [Moshchenko et al., 2019].

Based on metal and petroleum hydrocarbon contamination levels, bottom sediments of the Amur and Ussuri bays are classified as 'moderately polluted' or 'polluted' [Moshchenko et al., 2019]. The high spatial heterogeneity in pollutant distribution within bottom sediments of these bays, along with diverse grain size, results from the hydrodynamics in the region. The most contaminated areas are Zolotoy Rog Bay and Eastern Bosphorus Strait: there, concentrations of metals and hydrocarbons exceed by several times background values and ERL level (effects range-low, *i. e.*, the concentration level at which biological effects are unlikely [Long et al., 1995, cited after: Shulkin, 2004]). The Amur Bay exhibits moderate pollution level. The Ussuri Bay is the cleanest out of the studied areas. Researchers suggest that chemical pollution and eutrophication are currently key factors determining the ecological state of benthos off Vladivostok, and severe pollution of bottom sediments remains localized [Moshchenko et al., 2019].

Algae serve as integrative indicators of heavy metal pollution levels in coastal marine waters [Aboal et al., 2023; Khristoforova, 1989; Pan et al., 2018; Rainbow, Phillips, 1993; *etc.*]. Off Vladivostok, algae grow on rocky substrates in tidal and subtidal zones, primarily accumulating biologically available and highly mobile dissolved metals.

To identify the scale of environmental disturbance and level of metal pollution, researchers compare metal concentrations in macrophytes from studied areas with those from uncontaminated (background) sites [Obluchinskaya et al., 2013; Scanes, Roach, 1999]. However, selecting such background sites can be subjective. Alternatives include using background concentrations calculated as arithmetic or geometric means [Savenko, 2006], truncated means [Sánchez-Quiles et al., 2017], median values [Lukashev, 2007; Reimann et al., 2005; Sánchez-Quiles et al., 2017], or the 85th percentile [Cantillo, 1998]. Sanitary and hygienic MPC values for heavy metals (Hg, Pb, and Cd) and arsenic, used in Russia and abroad, are unsuitable for ecological assessments of natural systems. These standards were developed to determine the safety of food products and raw materials and not to reveal the well-being of hydrobionts themselves.

As already mentioned, to assess the ecological state of the marine environment at different stations off the Amur and Ussuri bays, we used threshold values of background metal concentrations ($C_{threshold}$ = median + 2 MAD) [Chernova, 2012; Chernova, Kozhenkova, 2016; Reimann et al., 2005] and hazard coefficients (K_H) calculated based on them (Figs 2, 3).

Iron and manganese compounds are of low toxicity to hydrobionts, as evidenced by high $MPC_{fishery}$ values for seawater (Table 3). Surface runoff is the primary source of dissolved Fe and Mn entering coastal waters of the bays from the Muravyov-Amursky Peninsula [Shul'kin, 2012]. In areas with high suspended matter content in seawater, metals accumulate in algae not only from the dissolved phase, but also from settling particulate matter [Burdin, Zolotukhina, 1998; Malinovskaya, Khristoforova, 1997]. The contribution of riverine input is maximum in the upper parts of the Amur and Ussuri bays adjacent to the western and eastern Muravyov-Amursky Peninsula, respectively (Fig. 1). There, elevated concentrations of these elements were noted in dissolved form (Table 3), in suspended form [Shulkin, 2004], and in algae (sta. 1, 2, and 27; see Figs 2, 3).

Bottom sediments are an even more indirect source of metals for macrophytes growing on hard substrates than suspended matter. In the Eastern Bosphorus Strait, where bottom sediments exhibit high levels of metal pollution [Moshchenko et al., 2019], elevated content of Fe, Mn, and Zn was also observed in algae (sta. 17 and 18). This is likely associated with the input of insufficiently treated wastewater from Vladivostok (domestic sewage and water from port infrastructure facilities), with shipping, and with large-scale sediment displacement during construction on the coast.

The Amur Bay experiences higher anthropogenic disturbances than the Ussuri Bay but lower ones than the Eastern Bosphorus Strait [Moshchenko et al., 2019; Vashchenko et al., 2010]. Typically, dissolved metal concentrations in seawater of these areas did not exceed $MPC_{fishery}$ [Marine Water Pollution, 2018], while heavy metal content in bottom sediments was elevated compared to permissible concentrations at which biological effects are unlikely [Losev, 2020; Marine Water Pollution, 2018; Petukhov et al., 2018; Vashchenko et al., 2010]. In total, metal concentrations in algae along the western Muravyov-Amursky Peninsula did not exceed thresholds. Fe, Mn, and Ni content was elevated in macrophytes from the shallow Uglovoy Cove (sta. 26 and 27) due to the inflow of several watercourses. Studies of metal and petroleum hydrocarbon pollution in bottom sediments of the Uglovoy Cove across different seasons indicate the bay's capacity for self-purification facilitated by catastrophic events like typhoons [Losev, 2020]. This contributes to the fact that metals do not accumulate in hydrobionts of the Amur Bay coastal waters at concentrations exceeding $C_{threshold}$.

Leachate both from the former Vladivostok landfill adjacent to the coastline and reclaimed in 2011 (sta. 6–9) and from ash ponds of the thermal power station No. 2 reclaimed before 2018 (sta. 10–12) is a significant anthropogenic source of contamination for coastal waters of the peninsula's eastern coast in the Ussuri Bay. High levels of metal pollution in marine ecosystem components in the landfill vicinity were noted both before its reclamation [Shulkin, 2004; Simokon, 2009] and after it. Importantly, after reclamation, the degree of marine environment pollution decreased [Belcheva et al., 2015; Kozhenkova et al., 2021]. Our results showed as follows: in summer 2017, Fe and Cu compounds were the key pollutants for algae from these stations, while Pb, Mn, Zn, Ni, and Cd were the secondary ones (co-pollutants) (Figs 2, 3). Comparison of dissolved metal concentrations in water (Table 3) with background levels [Shulkin et al., 2013] also revealed high levels of Cu (sta. 7), Zn, Cd, Ni, and Pb (sta. 6 and 7). In summer, leachate from the landfill spread predominantly northward due to prevailing southerly winds. Consequently, maximum metal concentrations in algae and water were observed not only at sta. 7, but also 2 km north, at sta. 6. However, under northerly and westerly winds, pollutants can be transported south and east thus increasing content of trace elements in water and algae at southern stations (sta. 8) (Fig. 2, Table 3). Metal concentrations in algae dropped sharply at the northern cape of the Desantnaya Cove (sta. 7) located 700 m from the southern one [Kozhenkova et al., 2021]. This fact aligns with data from [Chalkley et al., 2019] evidencing for the localized effect of pollution sources in water and a rapid decrease in metal concentrations in macrophytes to near-background levels within 100 m from a source.

Leachate from ash ponds of the thermal power station No. 2, entering Promezhutochnaya and Gornostay coves for many years, was another source of pollution for coastal waters of the western Ussuri Bay [Kozhenkova et al., 2021; Shulkin, 2004; Simokon, 2009]. However, in 2017, metal concentrations in algae at sta. 9–11 were comparable to those at sta. 12–16 within Vladivostok and generally did not exceed $C_{threshold}$ (Figs 2, 3). The levels of dissolved metals in water in this area (sta. 10) corresponded to background values, except for Zn (Table 3). The reclamation of ash ponds seems to be one of the reasons for the reduced input of polluted waters into the marine environment and decreased metal accumulation in water plants.

In the recreational zone of the Ussuri Bay (sta. 3–5), heavy metal concentrations in algae did not exceed $C_{threshold}$. Dissolved metal levels in water at the time of sampling also corresponded to background values for Peter the Great Bay (the Sea of Japan) (Table 3). Importantly, off the Russky and Popov islands (sta. 15, 16, and 20), Ni concentrations were elevated. This is likely associated with the heavy shipping traffic in the Eastern Bosphorus Strait and off Vladivostok in general. Shipping contributes to pollution of Peter the Great Bay waters with both petroleum hydrocarbons [Marine Water Pollution, 2018; Moshchenko et al., 2019] and nickel – a trace impurity in petroleum products [Yakubov et al., 2017].

High concentrations of dissolved metals in seawater in the landfill vicinity (sta. 7) are consistent with elevated content of trace elements in algae (Table 3, Figs 2, 3). However, according the Primorsky Territorial Office on Hydrometeorology and Environmental Monitoring, dissolved metal concentrations exceeding $MPC_{fishery}$ were not recorded in the Ussuri Bay in 2017, although the values registered were slightly higher than background ones [Marine Water Pollution, 2018]. Apparently, the discrepancy between our data and those of the Primorsky Territorial Office on Hydrometeorology and Environmental Monitoring is due to temporal differences and location of its sampling stations at deeper spots where mixing of leachate and seawater had already occurred. V. Shulkin [2004] also found dissolved metal concentrations in the landfill vicinity in 2001 that were elevated relative to background but did not exceed MPC. The elevated concentration of dissolved copper we detected in the landfill vicinity in 2017 (exceeding $MPC_{fishery}$) was likely related to the location of a sampling site (coastal zone, a depth of 1 m) and period of the study: before sampling, after a relatively dry June, there were more than 40 mm of precipitation in the first decade of July. Heavy precipitation leached mobile chemical elements from the 'body' of the landfill. Contaminated leachate was transported predominantly northward by a current and affected the chemical composition of bioindicators. As experimentally established, accumulation of metals by living algae from contaminated environments by 2 orders of magnitude occurs within 1.5–5 days [Suresh Kumar et al., 2007], while metal elimination occurs significantly slower and at a lower rate [Wang, Dei, 1999].

Metal concentrations in *Sargassum* spp. off the Muravyov-Amursky Peninsula were compared with generalized global indicators: the median and the third quartile (Q3) of element content in brown algae [Sánchez-Quiles et al., 2017]. Minimum Fe concentrations in *Sargassum* spp. from the Ussuri and Amur bays were found to practically correspond to the global median, while maximum ones exceeded Q3. Among Mn content values in *Sargassum* spp. from the study area, some were below the global median, but most concentrations exceeded Q3 (Table 2). The maximum Mn content in algae from the upper part of the Ussuri Bay, (5,063 ± 450) $\mu g \cdot g^{-1}$, was 4 times higher than the known Mn concentration in brown algae according to the compilation by Sánchez-Quiles *et al.* [2017]. We found the highest Mn content in *S. miyabei*, 5,863 $\mu g \cdot g^{-1}$, in the Abrek Cove, Strelok Bay, Sea of Japan [Kozhenkova et al., 2021]. In 2017, stations with Mn concentrations in *Sargassum* spp. exceeding Q3 were located in the upper part of the Ussuri Bay (sta. 1 and 2), where four small rivers inflow, and along the western coast of the bay (sta. 6 and 7). In other bays along the Russian coast of the Sea of Japan, Mn content exceeding Q3 in *Sargassum* spp. were characteristic of estuaries, ports, and spots used to dump dredged material from port operations [Chernova, Kozhenkova, 2016]. Concentrations of Cu, Zn, Pb, Cd, and Ni in *Sargassum* spp. from the study area did not exceed Q3 and were often below the median (Table 2).

Extremely high metal concentrations compared to global data were observed in a green alga *B. minima* at sta. 7, near the reclaimed solid waste landfill, off the western coast of the Ussuri Bay. Iron concentration in *Blidingia*, 38,813 $\mu g \cdot g^{-1}$, significantly exceeded the known maximum Fe value in *Enteromorpha compressa* from the Chilean coast, 23,000 $\mu g \cdot g^{-1}$ [Ratkevicius et al., 2003, cited after: Sánchez-Quiles et al., 2017]. Cu concentration, (601 ± 145) $\mu g \cdot g^{-1}$, was comparable to the known maximum, 750 $\mu g \cdot g^{-1}$ in *E. compressa* from the Chilean coast. Ni value in this alga, (48.6 ± 27.8) $\mu g \cdot g^{-1}$, was also close to the reported maximum, 83.4 $\mu g \cdot g^{-1}$ in *Halimeda tuna* from the Lebanese Mediterranean coast, Beirut [Shiber, Shatila, 1979, cited after: Sánchez-Quiles et al., 2017]. Concentrations of Zn and Pb in *B. minima* from sta. 7 were substantially higher than Q3 as well. Mn value did not exceed Q3. Cd concentration corresponded to the global median for green algae (Table 2).

Thus, manganese concentrations in brown algae off the Muravyov-Amursky Peninsula exhibited extremely high values compared to global literature data, while maximum concentrations of Cu, Zn, Pb, Cd, and Ni did not exceed the global Q3. Mn concentrations higher than Q3 in *Sargassum* spp. were characteristic of estuaries, ports, and spots used to dump dredged material. Maximum Fe and Cu values in green algae from the most polluted station (in the landfill vicinity) exceeded known literature values; Zn and Pb concentrations were higher than Q3; and Cd value corresponded to the global median for green algae.

There are several systems for the integrated assessment of chemical pollution using biological indicators [Richir, Gobert, 2014; Usero et al., 1996]. One of official systems (Andalusia, Spain) is the Metal Pollution Index, MPI [AMA, 1992, cited after: Usero et al., 1996]:

$$\mathrm{MPI} = (C_{f1} \times C_{f2} \times C_{f3} \times ... \times C_{fn})^{1/n} ,$$

where C_{f1}, C_{f2}… C_{fn} are concentrations of the 1st, 2nd… *n*-th element;

n is the number of elements analyzed in a sample.

J. Richir and S. Gobert [2014] used MPI [Usero et al., 1996] when assessing pollution by both metals and non-metals and named it TEPI. The authors emphasized the need for normalizing concentration of each element by its mean value in a dataset:

$$\mathrm{TEPI} = (Cf_1 \times Cf_2 \times ... \times Cf_n)^{1/n} ,$$

where Cf_1, Cf_2… Cf_n are concentrations normalized by their means (the ratio of a certain metal's concentration in the organism at a certain station to its mean value in the dataset);

n is the number of elements analyzed in a sample.

Normalization is useful when dealing with metals having concentrations that differ by orders of magnitude [Moreda-Piñeiro et al., 2001]. If metal concentration in macrophytes does not exceed the mean value, TEPI is ≤ 1. The higher TEPI, the higher the level of water pollution at the station. The authors [Richir, Gobert, 2014] assume that using this index allows for reliable comparison of global pollution levels across different monitoring periods, even if lists of analyzed elements and/or bioindicator species differ.

Notably, any integrated index averages the overall pollution level: an excess of one chemical element can be compensated by several others with concentration not exceeding threshold values. Specifically, the more metals with concentrations below mean values included in TEPI calculation, the lower (and closer to 1) the resulting calculated pollution level. The occurrence of a single substance with concentration significantly exceeding the regulatory standard can lead to a community distress [Risnik et al., 2012] and its restructuring.

When calculating TEPI for waters off the Muravyov-Amursky Peninsula, we used thresholds of background metal concentrations ($C_{threshold}$ = median + 2 MAD) for normalization instead of their mean values in the dataset (Fig. 4).

To avoid compensating for the excess of some elements (their values being higher than $C_{threshold}$) with others not exceeding thresholds, and to obtain a realistic indicator of metal pollution in macrophytes, TEPI-threshold was calculated only considering elements exceeding $C_{threshold}$ (Fig. 4). If none of the metals exceeded thresholds, one element with a concentration closest to the threshold was selected for TEPI determination.

Overall, the assessment of pollution levels in coastal areas off the Muravyov-Amursky Peninsula using integrated TEPI-threshold confirmed low pollution with heavy metals (Fig. 4). At half of the stations surveyed, TEPI-threshold was less than 1 or equal to it. At other sites, index values ranged 1.1–3.2; this was primarily due to metal input into the coastal environment *via* riverine discharge into the upper part of the Amur and Ussuri bays. The maximum metal pollution off the peninsula was localized near the reclaimed solid waste landfill (TEPI-threshold = 16).

Conclusions. In July 2017, according to hazard coefficients (K_H) for individual elements and the integrated Trace Element Pollution Index (TEPI-threshold) for algae, coastal waters off Vladivostok exhibited low pollution with trace elements. A localized zone of high contamination formed by surface runoff of pollutants from the territory of the solid waste landfill in Vladivostok (TEPI-threshold = 16) was situated off the eastern coast of the Muravyov-Amursky Peninsula. Out of the elements studied, Fe and Cu were the primary pollutants for macrophytes here ($K_H > 80$), while Pb, Mn, Zn, and Ni were co-pollutants. Values of dissolved metals in seawater at this station exceeded background levels, with copper concentration reaching 3 MPC for fishery reservoirs. Concentrations of dissolved metals in seawater at other stations at the time of sampling generally corresponded to background values for Peter the Great Bay, Sea of Japan, except for Zn.

TEPI-threshold at stations bordering the solid waste landfill to the north and south ranged 2.4–2.8 mostly due to Pb and Cu pollution (2.7–12 K_H), but also due to Zn, Fe, Mn, and Ni contamination. Algae from the upper parts of the Ussuri and Amur bays were Fe- and Mn-enriched because of riverine metal input; TEPI-threshold was 1.7–3.0. In the Eastern Bosphorus Strait, elevated levels of Fe (3–10 K_H), as well as Mn, Cu, Zn, and Ni (1–1.5 K_H), were noted in macrophytes. This was associated with port activities, shipping, and construction; TEPI-threshold was 1.0–2.1.

When calculating TEPI-threshold, normalizing by $C_{threshold}$ and including only elements with content equal to threshold values of background concentrations or exceeding it allow for the most objective assessment of the pollution level of the water area and prevent compensation for the excess of some elements by others not exceeding background levels.

In brown algae (*Sargassum* spp.) off the Muravyov-Amursky Peninsula, extremely high Mn concentrations compared to global data were found. In green algae, Fe and Cu values exceeded natural levels known in literature.

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ОЦЕНКА ЗАГРЯЗНЕНИЯ ТЯЖЁЛЫМИ МЕТАЛЛАМИ ПРИБРЕЖНЫХ ВОД ПОЛУОСТРОВА МУРАВЬЁВА-АМУРСКОГО С ИСПОЛЬЗОВАНИЕМ ВОДОРОСЛЕЙ-БИОИНДИКАТОРОВ

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Изучено содержание Fe, Mn, Cu, Zn, Pb, Cd и Ni в бурых водорослях [*Sargassum miyabei* Yendo и *S. pallidum* (Turner) C. Agardh] и зелёных водорослях [*Blidingia minima* (Nägeli ex Kützing) Kylin, *Ulva lactuca* Linnaeus и *U. linza* Linnaeus] из прибрежных вод полуострова Муравьёва-Амурского Японского моря, в окрестностях города Владивостока, собранных в июле 2017 г. Концентрации тяжёлых металлов в водорослях устанавливали методом атомно-абсорбционной спектрофотометрии после минерализации талломов с помощью азотной кислоты. Содержание растворённых элементов в морской воде определяли атомно-абсорбционным методом после ультрафильтрации проб воды и концентрирования металлов с помощью системы хлороформ — ДДТК-Na. На основе коэффициента опасности загрязнения водорослей металлами (K_O), представляющего собой отношение концентрации металла в водоросли к верхнему пороговому уровню фоновых концентраций элемента, а также на основе интегрального коэффициента TEPI-порог, рассчитанного с использованием $K_O \geq 1$, провели оценку степени загрязнения различных участков прибрежной зоны моря. Прибрежные воды вокруг Владивостока были слабо загрязнены тяжёлыми металлами. Индекс TEPI-порог на станциях к северу и югу от полигона твёрдых бытовых отходов (ТБО) составил 2,4–2,8 в связи с загрязнением Pb и Cu (2,7–12 $C_{порог}$), а также Zn, Fe, Mn и Ni. Водоросли из вершин Уссурийского и Амурского заливов обогащены Fe и Mn из-за выноса металлов водами рек; TEPI-порог — 1,7–3,0. В проливе Босфор Восточный загрязнение макрофитов Fe (3–10 $C_{порог}$), а также Mn, Cu, Zn и Ni (1–1,5 $C_{порог}$) связано с портовой деятельностью, судоходством и строительством мостов; значение TEPI-порог составило 1,0–2,1. Локальная зона высокой степени загрязнения, сформированная за счёт дренирования дождевыми водами рекультивированного полигона ТБО города Владивостока, с TEPI-порог = 16, находится у восточного побережья полуострова Муравьёва-Амурского. Из числа исследованных элементов Fe и Cu были основными загрязнителями макрофитов этой станции ($K_O > 80$ в водорослях), а Pb, Mn, Zn и Ni — сопутствующими. В морской воде с этой станции концентрации растворённых металлов превышали фоновые уровни, содержание растворённой меди составляло 3 ПДК для рыбохозяйственных водоёмов.

Ключевые слова: загрязнение, тяжёлые металлы, бурые водоросли, зелёные водоросли, Амурский залив, Уссурийский залив, залив Петра Великого, полуостров Муравьёва-Амурского, Японское море