

Assessment of Heavy Metal Pollution of the Peter the Great Bay (North-West Pacific Region) Using Brown Algae

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Abstract

According to the content of heavy metals Fe, Mn, Cu, Zn, Pb, Cd and Ni in brown algae *Sargassum miyabei* from the Peter the Great Bay of the Sea of Japan, geochemical factors were calculated. The algae geochemical anomaly index (I_{GA}) characterizes the degree of excess of the background concentrations of metals. The heavy metal pollution factor (F_p) is used to estimate the degree of pollution of the marine environment with heavy metals. The Peter the Great Bay has a low level of heavy metal pollution, but some parts of second-order bays, such as Amurskii Bay, Ussuriiskii Bay and Vostok Bay, have a moderate degree of pollution. The high pollution level was registered on the western coast of the Ussuriiskii Bay near the Vladivostok city landfill. Thirty percent of sampling stations were noted to have higher threshold levels of metals in the algae. These areas need to be monitored to assess their environmental status and measures should be applied to reduce the impact on the environment.

Keywords

Biomonitoring, Heavy Metals, Environmental Quality Regulation, Brown Algae, *Sargassum miyabei*, North-West Pacific Region

1. Introduction

The heavy-metals pollution of the marine environment is the pressing problem. The basis line in the solution of this problem is a decrease in income of contaminants from the main sources of pollution as well as a getting the positive balance between assimilatory capacity of the coastal ecosystem and intensity of the pollution supply (Israel, 1989). Monitoring of the environment is a basis for base

for making the specific decisions related to quality control of the coastal biogeosystems. The correct assessment of the degree of metal pollution in coastal areas should include the state analysis of different components of the ecosystem including the abiotic (water and bottom sediments) and biotic ones (Burdin, 1985; Khristoforova, 1989; Rainbow & Phillips, 1993; Chakraborty, Bhattacharya, Singh, & Maity, 2014).

Brown algae as the accumulative bioindicators are widely used to determine the degree of heavy-metals pollution of the coastal seawaters. The European researchers were the first to use brown algae for the ecological monitoring during the early 1970s (Bryan & Hummerstone, 1973; Fuge & James, 1974). Gradually this approach came into use in different regions of the world (Vasquez & Guerra, 1996; Amado-Filho, Salgado, Rebelo, Rezende, Karez, & Pfeiffer, 2008).

The research of the heavy metal concentrations in brown algae of the North west Pacific started in 1976 (Khristoforova, Bogdanova, & Tolstova, 1983; Khristoforova, 1989). Several species of algae species (*Costaria costata*, *Fucus evanescens*, *Saccharina japonica*, *Scytosyphon lomentaria*, *Silvetia babingtonii*, *Stephanocystis crassipes*) were used. Species and age features of the microelement composition of plants under the impact and background conditions were carried out, and the long-term estimates of the metal pollution were performed (Kozhenkova, Khristoforova, & Chernova, 2000; Shulkin, Chernova, Khristoforova, & Kozhenkova, 2015).

Two species of the *Sargassum* genus—*Sargassum miyabei* and *S. pallidum*—are widely distributed along the Russian coast of the Sea of Japan and to the south of it. For the first time, these species were used for assessment of heavy metal pollution of the coastal waters in the North west Pacific in late 1970s-early 1980s (Khristoforova, Bogdanova, & Tolstova, 1983).

Hereupon, different species of *Sargassum* genus were used as the heavy metal pollution indicators near Vietnam and Brasilia as well as in the East China Sea, Mediterranean Sea and other seas (Ho, 1988; Hou & Yan, 1998; Chernova, Khristoforova, & Vyshkvartsev, 2002; Al-Masri, Mamish, & Budier, 2003; Chernova & Sergeeva, 2008; Brito, de Souza, Bressy, Moura, & Korn, 2012; Chernova & Khristoforova, 2012; Sun, Liu, Jiang, & Yang, 2019).

The purpose of the article is to assess the heavy metal pollution of the Peter the Great Bay (PGB) with brown alga *Sargassum miyabei*.

2. Study Area

The Peter the Great Bay is located in the northwestern Sea of Japan between 42°17'N and 43°20'N, 130°41'E and 133°02'E. The Bay area exceeds 6000 km². According to the type of the coastal line and bottom relief, the Bay is divided into several parts.

The southwestern part of the Bay is located between the estuary of the Tumen River in the south and the Cape of Bruce in the north. The Far-Eastern State

Marine Reserve (FESMR) occupies a part of the water area and coastal zone adjacent to it. The Muravyov-Amursky Peninsula, Russki, Popov Islands and a great number of small islands divide the northern Peter the Great Bay into two water areas—Amurskii Bay and Ussuriiskii Bay. Vladivostok city is located in the Muravyov-Amursky Peninsula and Russkii Island. The Strelok, Vostok and Nakhodka Bays are situated further east.

The Tumen River flows into the Sea of Japan on the south boundary of the Peter the Great Bay. Predominance of the southward Primorski Current causes a transportation of the river discharge to the shores of the Korean Peninsula and reigns in the influence of the Tumen River flow on the Russian coastal waters (Shulkin & Semykina, 2012). The Razdolnaya, Partizanskaya Rivers and a great number of small rivers flow into the PGB.

The total area of the Peter the Great Bay catchment basin is 23,800 km². About a half of this territory is occupied by forests (10,095 km²) and protected natural areas (1157 km²).

Within the PGB catchment basin reside, 1.3 million people. Here, the cities of Vladivostok (633 thou. people), Ussuriisk (194 thou. people), Artem (114 thou. people), Fokino (31 thou. people), Bolshoi Kamen (40 thou. people), Nakhodka (154 thou. people) and Partizansk (45 thou. people) are situated (Brief..., 2016).

Power, machinery production, woodworking, chemical, textile, food industries, construction and agriculture are basic industries on the territory of the PGB catchment basin. The amount of wastewaters produced within the catchment basin has reduced from 620 million tons in 1990 to 290 million tons in 2015 (Brief..., 2016). The main reason of the wastewater amount reduction is a decline in the industrial production since 1990s.

3. Materials and Methods

The brown alga *Sargassum miyabei* was collected in July-August in 1995, 1998, 2000-2004, 2008, 2010 and 2017, with varying periodicity at 65 stations in the Peter the Great Bay as well as at two stations in the Kievka Bight located north-east of this Bay. The preparation of samples for the chemical analysis was earlier described (Chernova & Kozhenkova, 2016). The concentrations of Fe, Mn, Cu, Zn, Pb, Cd and Ni in the macrophytes were determined by the atomic absorption spectrophotometry method at Shimadzu 6800 in the Common Use Centre “Center of the Landscape Ecodiagnosics and GIS-technologies” in the Pacific Geographical Institute FEB RAS.

Concentrations of Fe, Mn, Cu, Zn and Pb were used for calculation of the geochemical coefficients: algae geochemical anomaly index (I_{GA}) and heavy metal pollution factor (F_p) (Figure 1). In case of absence Pb concentrations, Cd concentrations were used.

Apart from the author’s data, the published concentrations of metals in *S. miyabei* from the coastal zone of the Amurskii Bay (2011), Nakhodka Bay

Algae-geochemical-anomaly-index (I_{GA}): α	Heavy-metal-pollution-factor (F_p): α
$I_{GA} = \frac{1}{N} \sum_{i=1}^N \frac{C_i}{C_{med}}$	$F_p = \frac{1}{N} \sum_{i=1}^N \frac{C_i a_i}{C_{UT}}$

Figure 1. Formulas of the geochemical coefficients: I_{GA} —algae geochemical anomaly index, and F_p —heavy metal pollution factor. Notations: C_i is concentration of i -metal in *S. miyabei*, C_{med} is the background concentration of metal which is equal to median of metal concentrations in the shared dataset for *S. miyabei* from the north-western Sea of Japan (Kozhenkova & Chernova, 2018), N is the number of elements used for index, C_{UT} is the threshold concentration of metal for this type of algae calculated earlier (Chernova & Kozhenkova, 2016) as a median plus double median of absolute deviations from median ($Me + 2MAD$); a_i is the danger coefficient of i -element depending on danger class of element: $a = 2.5$ in case of I danger class, $a = 1.5$ in case of II danger class (Cd, Pb), $a = 1.0$ in case of III danger class (Cu), $a = 0.5$ in case of IV danger class (Fe, Mn).

(2012), south-western part of the Peter the Great Bay and around Russki Island (2016) were used (Kobzar & Kristoforova, 2015; Khristoforova, Gamayunova, & Afanasyev, 2015; Khristoforova & Kobzar, 2017; Khristoforova, Emelyanov, & Efimov, 2018). The stations of macrophyte sampling by the authors of this paper and our colleagues have intersected and, thereby, a total number of points of macrophyte sampling in the Peter the Great Bay reaches 84 stations.

The calculations of F_p for some second-order bays which are the parts of the Peter the Great Bay were performed on the basis of all data over the years and at all stations. The common value of F_p for the Peter the Great Bay was calculated after exclusion from shared dataset the values exceeding the percentile P_{85} . The calculation of the mean, median and percentiles (P_{25} , P_{75} , P_{85}) was performed with Excel. Based on the mean values of F_p , degree of environmental pollution was assessed as **light** if $F_p = 0.4 - 0.5$, or as **low** if $F_p = 0.51 - 0.8$, or as **moderate** if $F_p = 0.81 - 3.5$, or as **high** if $F_p = 3.51 - 7.5$, or as **heavy pollution** if $F_p > 7.5$ (Kozhenkova & Chernova, 2018).

4. Results

In total I_{GA} varied in a range of 0.3 - 7.5, and 50% of values (percentile 0.25 - 0.75) changed within the limits of 0.8 - 1.5. Based on heavy metal concentrations in *S. miyabei*, I_{GA} was lesser than 1 for 56% of stations in the Peter the Great Bay, and the values of the geochemical anomaly coefficient exceeded 2 at 20% of stations. These are stations in the Amurskii Bay: Sanatornaya (1995, $I_{GA} = 5.1$) at the expense of contribution of Mn, Fe, Pb, Zn and Cu (in descending order of C_i/C_{med} , here and further), Tokarevsky Cape (1995, $I_{GA} = 4.0$; 2002, $I_{GA} = 2.7$; Mn, Pb, Fe, Zn and Cu), Markovsky Cape (2011, $I_{GA} = 3.5$) at the expense of contribution of Pb and Cu, Pionerskaya Bight (1995, $I_{GA} = 3.3$; Mn and Cu) and other stations situated along the coast of the Muravyov-Amursky Peninsula where the Vladivostok city is situated. In the Ussuriiskii Bay, the largest I_{GA} was found for the Desantnaya Bight (7.5 in 2016 and 5.6 in 2017) at the expense of Cu, Pb and

Mn contribution the source of which is the Vladivostok landfill. In Strelok, Vostok and Nakhodka Bays, the stations with $I_{GA} > 2$ were also revealed. At the same time, the values of I_{GA} at stations in Kievka Bight and south-western Peter the Great Bay were not increased.

Heavy metal pollution factor (F_p) at different stations in PGB coast has varied in a range of 0.2 - 4.7, a percentile P_{85} was equal to 1.0, thus the mean value of $F_p = 0.55$. This characterizes the degree of pollution as low.

The water pollution in different parts of the Bay varies from light (south-western part) to moderate one (Amurskii Bay and Ussuriiskii Bay) (**Table 1**).

In the Amurskii, Ussuriiskii and Vostok Bays the points with moderate degree of heavy-metals pollution ($F_p > 0.80$) were identified (**Table 2, Figure 2**). The

Table 1. Heavy metal pollution factor (F_p) for different parts of the Peter the Great Bay.

Part of the PGB	Median	Mean \pm SD	Range	Water pollution
South-West	0.45	0.49 \pm 0.18	0.49 - 0.79	light pollution
Amurskii Bay	0.65	0.85 \pm 0.51	0.34 - 2.67	moderate pollution
Ussuriiskii Bay	0.62	1.03 \pm 1.06	0.35 - 4.65	moderate pollution
Vostok Bay	0.43	0.78 \pm 0.72	0.2 - 3.13	low pollution
Nakhodka Bay	0.52	0.52 \pm 0.19	0.26 - 1.17	low pollution
Kievka Bight	0.40	0.36 \pm 0.14	0.15 - 0.51	pure

Table 2. Heavy metal pollution factor (F_p) for polluted locations in Ussuriiskii and Vostok Bays.

Year	Location	F_p	Water pollution	Pollutants
Ussuriiskii Bay				
2004	Gornostay Bight	1.30	moderate pollution	Cu, Zn, Pb
2017		0.59	low pollution	
2016	Desantnaya Bight	4.65	high pollution	Cu, Zn, Pb, Fe, Mn
2017		3.17	moderate pollution	
2002	2 km to the north from the Desantnaya Bight	1.95	moderate pollution	Cu, Pb, Ni
2010	Muravinaya Bight	0.85	moderate pollution	Fe, Mn, Ni, Cd
2017		1.22	moderate pollution	
2010	Telakovskogo Bight	0.89	moderate pollution	Fe, Mn, Ni
Vostok Bay				
2002	Gaydamak Bight	1.18	moderate pollution	Cu, Zn, Pb, Mn
2003		1.18	moderate pollution	
2004		0.78	low pollution	
2002	Srednaya Bight	0.94	moderate pollution	Zn, Pb
2002	Tihaya Bight	2.25	moderate pollution	Cu, Zn, Pb
2002	Noth-western part of the Bay	0.85	moderate pollution	Fe, Mn
2002	Northern part of the Bay	1.12	moderate pollution	Fe, Mn

high pollution level was determined on the western coast of the Ussuriiskii Bay near the Vladivostok city landfill ($F_p > 3.50$). The total number of sampling stations with excess of the threshold levels of heavy metals in *S. miyabei* reached 24, thus, based on the degree of pollution, 30% of inspection sites are characterized by moderate or higher pollution.

A long-term monitoring of the heavy metal pollution degree based on F_p in 1995-1998, 2000-2005, 2008-2012 and 2015-2017 was carried out for the following three areas of the Peter the Great Bay: south-western part, Amurskii Bay and Nakhodka Bay (**Figure 3**). A value of F_p for the south-western part of the Bay varied from 0.55 ± 0.19 in 2000-2005 to 0.34 ± 0.09 in 2015-2017 (low pollution → pure), in Amurskii Bay from 0.95 ± 0.49 in 1995-1998 to 0.65 ± 0.37 in 2015-2017 (moderate pollution → low pollution), and in Nakhodka Bay from 0.54 ± 0.36 in 1995-1998 to 0.50 ± 0.12 in 2008-2012 (low pollution → light pollution).

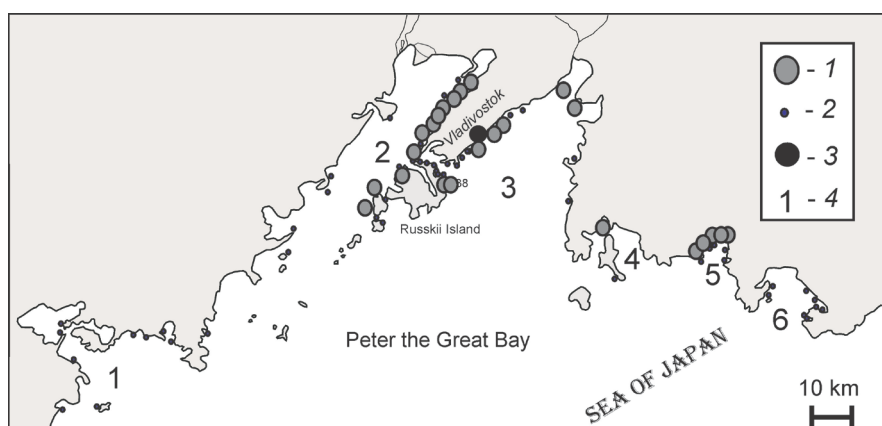


Figure 2. Heavy metal pollution in the Peter the Great Bay, Sea of Japan: 1—locations with $F_p > 0.8$; 2—locations with $F_p \leq 0.8$, 3—Vladivostok city landfill, 4—parts of PGB (1—south-western part, 2—Amurskii Bay, 3—Ussuriiskii Bay, 4—Strelok Bay, 5—Vostok Bay, 6—Nakhodka Bay).

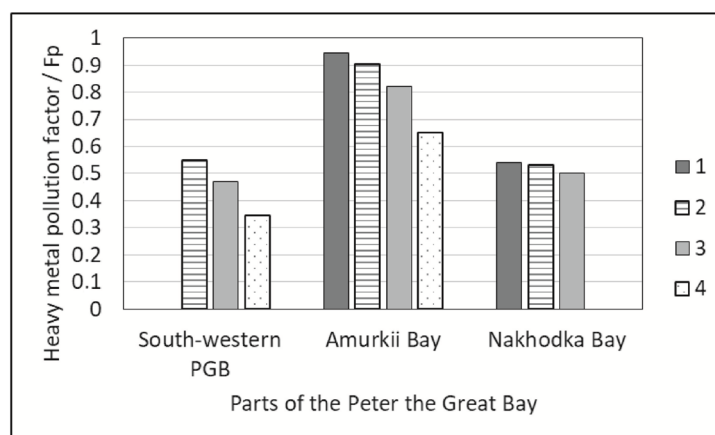


Figure 3. Change in the heavy metal pollution degree in different parts of the Peter the Great Bay at the end of XX—at the beginning of XXI centuries: 1) 1995-1998, 2) 2000-2005, 3) 2008-2012, 4) 2015-2017.

5. Discussion

The determination of the space structure of pollution differentiating the territory with respect to level of ecological hazard is the crucial task of the geochemical surveys of environment.

The main routes of metals to the marine environment are river runoff and atmospheric deposition (Shulkin, 2012). The anthropogenic sources of pollutants such as sewage waters of manufacturing facilities and residential areas, land drainage from the urbanized terrains and municipal solid waste landfills, corrosion of the facilities of port infrastructure and ship bottoms, sunken floating crafts and some other objects change the natural geochemical flows of elements.

The different parts of the Peter the Great Bay are characterized by different sources of pollutants to the sea. In 1980s-1990s, the degree of anthropogenic impact and volume of entry of the organic and inorganic pollutants to different parts of the Bay fell in the following sequence: Amurskii Bay > Ussuriiskii Bay > Nakhodka Bay > Strelok Bay > Vostok Bay, Posyet Bay > FESMR (Ogorodnikova, 2001; Shulkin, 2004). Decline in the volumes of industrial and agricultural outputs in the late 1990s resulted in decrease of pollutants entry to the coastal seawaters (Lukyanova, Cherkashin, & Simokon, 2012).

Determination of the pollution degree of the environment by different chemicals is based on the principle of comparison of their actual concentrations in water, bottom sediments, soil or air with accepted ecological standards such as maximum permissible concentrations (Guidelines..., 1988; Shiriev, 2005). The concentrations of metals in the abundant species of marine organisms—bivalves, brown and green algae—are also used as indicators of the marine environment state (Amado-Filho, Salgado, Rebelo, Rezende, Karez, & Pfeiffer, 2008; Pan, Wernberg, de Bettignies, Holmer, Li, Wu et al., 2018; Sun, Liu, Jiang, & Yang, 2019) throughout the comparison with the background values. Maximum permissible concentrations for organisms were not determined yet, but there are several approaches to their determination (Cantillo, 1998; Reinmann, Filzmoser, & Garrett, 2005; Lukashev, 2007; Zalewska & Danowska, 2017). In many countries, the health-based exposure limits were approved for determining the safety in use of bioresources but they are not in line with goals of maintaining the stability of ecosystems.

The degree of environmental hazard of territory and water area can be determined using the different kinds of combined and/or integrated pollution indices. The international Metal Pollution Index (MPI) proposed by J. Usero with coauthors (1996), in its current form, only allows to compare the global trace metal pollution between the different monitored sites of a specific survey, for a given species (Richir & Gobert, 2014). Hereafter, two new adapted indices—TESVI (Trace Element Spatial Variation Index) and TEPI (Trace Element Pollution Index)—which were based on account of the statistically average values—mean standard deviation, maximum and minimum values of metal concentrations in a dataset were proposed (Richir & Gobert, 2014).

Based on the study of C. Reinmann with coauthors (2005), we have calculated the background and threshold heavy metal concentrations in algae of the coastal part of the Sea of Japan (Chernova, 2012; Chernova & Kozhenkova, 2016), as well as for Okhotsk Sea, White Sea and South China Sea (Chernova & Khristoforova, 2012).

Brown algae are the integral indicators of heavy metal pollution in the coastal waters (Rainbow & Phillips, 1993; Vasquez & Guerra, 1996; Shulkin, Chernova, Khristoforova, & Kozhenkova, 2015; Chernova & Kozhenkova, 2016; Pan, Wernberg, de Bettignies, Holmer, Li, Wu et al., 2018). Biogeochemical coefficients reflect the features of the microelement composition of the brown alga *S. miyabei* in the different localities or within a “separate” water area (**Table 1, Table 2**). A value of I_{GA} (*algae geochemical anomaly index*) assesses the features of the content of the metal complex in algae with respect to the background conditions. As the background concentration, the median of the dataset with the excluded statistical outliers was used. Therefore, $I_{GA} = 1$ corresponds to the background level of metals in the alga. If a value of $I_{GA} < 2$, the algae grow in the environment with natural level of metal content (Chernova & Kozhenkova, 2020). However, at $I_{GA} > 2$, the anthropogenic effect on the entry of metals to the marine environment becomes deciding. In the Peter the Great Bay, the stations with $I_{GA} > 2$ are located on the eastern coast of the Amurskii Bay and western coast of the Ussuriiskii Bay and related to heavy metal pollution from the industrial and municipal sewage waters of Vladivostok, washout of pollutants on the town landfill territory and effect of the port. In the Strelak, Vostok and Nakhodka Bays, the stations with $I_{GA} > 2$ are located near ports and outlets of polluted wastewaters from populated areas.

Different metals are characterized by different toxicities for the marine organisms (Chu & Chow, 2002; Golovanova, 2008; Mebane, Hennessy, & Dillon, 2008). Heavy metal pollution factor (F_p) characterizes the enrichment of algae with heavy metals with respect to the threshold levels and with consideration for the hazard class for marine organisms. All the chemical substances in the fishery water bodies were separated into 4 hazard classes (Fe, Mn—4; Cu, Zn, Ni—3; Cd, Pb—2; Order..., 2016). The stations with the largest values of F_p are rated as most dangerous from the toxicological standpoint for the marine biota. Therefore, the measures related to reducing the volumes of metal entry to the coastal seawaters in such areas should be carried out in the first place.

According to F_p value for *S. miyabei* the pollution degree of PGB water areas is rated as low. Different parts of the second-order bays, such as Amurskii, Ussuriiskii and Vostok Bays, have a moderate degree of pollution (**Figure 2, Table 2**). The high pollution level was determined on the western coast of the Ussuriiskii Bay near the Vladivostok city landfill (**Table 2**).

Our conclusions of the degree of the Peter the Great Bay pollution with heavy metals confirm and supplement the data on monitoring of this Bay pollution with the use of the bottom sediments (BS). Generally, the content of metals in

the BS of the Bay characterizes the ecosystem state as advantageous (Dudarev, Botsul, Charkin, Biryulina, & Gavrilova, 2002). Bottom sediments within the Port of Vladivostok, i.e. in the Golden Horn Bay and Diomid Bight, are heavily polluted by metals. The concentrations of lead, cadmium, mercury, zinc and copper in the BSs of these water areas exceed 10 and more times the background level (Shulkin, 2004). The brown algae *S. miyabei* were not found within the Port of Vladivostok, so we have not data on heavy metal concentration in the macroalgae from this area. In the south-eastern part of the Amurskii Bay, a zone with concentrations of Pb, Cd, Cu and Zn in BS exceeding the background values 1.5 - 3 times has formed which is probably caused by dumping of contaminated sediments from the Port of Vladivostok. In the south-western coast of the Ussuriiskii Bay the BS contain Pb, Cu and Zn with concentrations higher 2 - 3 times than the background levels. The maximal metal pollution of sediments was recorded near the municipal landfill on the western coast of the Ussuriiskii Bay: more than 13-fold excess of the background for Pb, Cd, Hg and Cu (Lukyanova, Cherkashin, & Simokon, 2012; Shulkin & Semykina, 2012).

Reduction in the volumes of industrial and agricultural production in the south of the Russian Far East in the late 1990s has resulted in decreasing the volumes of pollutants (Brief..., 2016). The tendency to reducing the degree of heavy metal pollution in the Peter the Great Bay for the period from the end of the XX century to early XXI century was revealed on the basis of F_p values for brown alga *S. miyabei* (Figure 3, Table 2). A lowering in content of different pollutants in the sea water and bottom sediments of PGB was also recorded. The ecotoxicological situation in the most polluted water areas improved (Cherkashin, Simokon, & Pryazhevskaya, 2019). It has been established that the state of the benthos and fishes community was stable. The ichthyofauna of the Peter the Great Bay remains rich in species composition and the density of fish concentration is high here. Biomass of fishes is little different in the Amurskii (11.6 t/km²) and Ussuriiskii (12.2 t/km²) Bays as well as in the open Peter the Great Bay. In the Amurskii Bay, the effect of pollution from the sewer outfalls is only recorded in the north-eastern part of the Bay where the harvestings of many species of fishes, especially flounders, became smaller (Lukyanova, Cherkashin, & Simokon, 2012).

Over the last years, a growth of oil transportation and volume of agricultural production are observed in the southern Far East of Russia. It can increase the input of pollutants including heavy metals to the catchment basin of the Peter the Great Bay. For this reason, the monitoring of marine environment should be continued.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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