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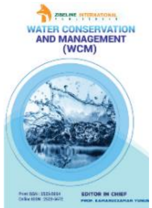

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RESEARCH ARTICLE

INFLUENCE OF ANTHROPOGENIC IMPACT ON THE PROCESS OF MANGANESE RELEASE FROM THE SEDIMENTARY STATE UNDER THE ACTION OF POLLUTANTS WITH REDUCING ACTIVITY IN THE CHERNAYA RIVER (Khabarovsk)

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ABSTRACT

The article presents the results of the study of seasonal and spatial variations of manganese content in the waters of small rivers in urbanized areas of the south of the Russian Far East using the example of the Chernaya River draining the vicinity of Khabarovsk. The anthropogenic impact leads to the fact that in surface waters reducing conditions are formed, leading to the release significant amounts of manganese (more than 1000μg/l). Taking into account the absence of anthropogenic sources of manganese in this area, it can be assumed that manganese is released from the products of hydrolytic degradation of ferromanganese nodules. It is shown that the process is most intensive in winter. Wastewater pollutants, responsible for the increase in permanganate index, when increasing permanganate index by every 10 mg(O_2)/l lead to the release of 83 μg Mn (summer period) to 387 μg Mn (winter period). The maximum recorded content at this time reaches 1198 μg/l Mn.

KEYWORDS

Manganese, surface water, ferromanganese nodule,

1. INTRODUCTION

In the Earth's crust, manganese ranks third (after iron and titanium) among the most abundant transition metals (Armstrong, 2008). Chronic exposure to toxic doses of manganese is called manganism. This condition was first described by John Cooper in 1837 in two workers in Scotland who were involved in the process of grinding manganese ore. As a result of such work, they developed symptoms of paraplegia (Furbee and Dobbs, 2009). In toxic doses, manganese exhibits the properties of a polytropic poison, affecting lung tissue, cardiovascular and hepatobiliary systems, while aggravating the course of allergic processes. Manganese can also have mutagenic activity, and its excessive content in drinking water has a toxic effect on the pancreas, liver and can cause skin diseases (Ying et al., 2017). The World Health Organization has set a maximum manganese concentration of 0.1 mg/l for drinking water supply systems (Usepa, 2004).

Manganese is an element with variable valence. In natural biogeocenoses and geological formations, as a rule, there are conditions leading to the formation of manganese (II) and manganese (IV) compounds (Ma et al., 2023). Recently, some researchers have paid additional attention to manganese (III) compounds (Shi et al., 2020; Oldham et al., 2017), which are metastable in aqueous solutions but relatively stable when incorporated into minerals. At low concentrations, manganese is an essential trace element and is assimilated by aquatic organisms mainly in the ionic form Mn^{2+}. However, at a concentration of 9.4 mg/L, manganese can be a toxic element with an LC50 in 48 h (lethal concentration), e.g. for *Daphnia magna* (Howe et al., 2004). Manganese (II) enters the body mainly with food and drinking water, so its regular monitoring in such objects is important for human safety (Crapnell and Banks, 2022).

Manganese occurs naturally in many surface waters, groundwater, and soils. Human activities in some areas are also responsible for significant manganese contamination of waters. As mentioned earlier, ingestion of manganese through food and drinking water is the main route of exposure to manganese in the general population. Frisby and colleagues (Frisbie, 2012) reported that drinking water with Mn concentrations greater than 400 μg/L was found in 54 countries worldwide, affecting tens of millions of people.

As experimental studies and computer modeling show, dissolved manganese is very mobile in the cationic form Mn^{2+}. In this state, it is less prone to form chelate compounds compared to other trace elements such as Cu, Co, Ni, Pb or Zn (Abbasse et al., 2022; Charriau et al., 2011; Morgan, 2000). As a consequence, manganese in the soluble form is able to remain in the mobile state for a relatively long time and to be transported in water bodies for considerable distances.

Manganese is an example of a metal for which the influence of the content of organic pollutants on its migration activity is particularly significant. This is due to the fact that it actively interacts with the organic component of pollution, participating in redox hydrochemical processes. In this case, it is released from bottom sediments and transformed into the mobile ionic form Mn^{2+}. When water is saturated with oxygen, the opposite process takes place. Manganese undergoes oxidation and transforms into insoluble form MnO_2, turning out to be a part of bottom sediments. Significantly, the dissolved oxygen content presents a spatial gradient change (Kaufman et al., 2017, Mader et al., 2018), inducing complex dynamic biogeochemical processes (Meng et al., 2020, Zhu et al., 2020). These processes will change the redox conditions and nutrient distribution, impacting the biotic and

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abiotic transformation of various elements and compounds (Feng et al., 2023).

Fresh waters in the south of the Russian Far East are characterized by elevated background values of manganese. This is due to the fact that the geochemical peculiarity of this region is the presence of ferromanganese nodules confined to soils and the lower part of the Pliocene clay strata. Accordingly, Khabarovsk and its vicinity are located in the territory of the so-called province of ferromanganese fresh waters (Kulakov, 2013; Novikov et al., 2008). Up to 5000-8000 μg/l of manganese is registered in groundwater not infrequently. This is a significant problem for water treatment of such waters for drinking use (Kulakov 2013).

Most of the literature data on the mobility of various forms of manganese and its migration in watercourses describe the behavior of this element for much smaller concentration ranges than those given above. For example, the upper limit of manganese content in the waters of European rivers rarely exceeds 40 μg/l (Superville et al., 2018). As a result, the conclusions about manganese mobility obtained in such studies cannot be correctly transferred to the concentration range characteristic of Outer Manchuria waters. Unfortunately, to date, the level of study of manganese in surface waters in the territories with ferromanganese nodules is not high enough. In general, articles are limited to the study of groundwater only, where the conditions of hydrochemical processes differ significantly from surface water.

Thus, new migration pathways of heavy metals due to anthropogenic influence are formed in urbanized areas. The study is focused on a comprehensive study of migration pathways of manganese and biogenic elements, as well as identification of additional migration pathways due to anthropogenic-initiated processes in rivers draining urbanized areas.
The objective of this study was to investigate the spatial and temporal release of manganese in watercourses of urbanized areas in the presence of ferromanganese nodules.

2. Material And Methods

2.1 Study Area

The Chernaya River drains the territory of Khabarovsk city and its surroundings (Figure 1, Table). This watercourse is characterized by a more significant length compared to other small rivers draining the area. The area of the river basin is 255km2. The area of residential area is 7.1%, industrial land - 6.7%, agricultural land - 48.4%, conditional natural land - 37.8%. The Chernaya River is subject to various anthropogenic impacts, but it does not receive industrial wastewater carrying elevated levels of heavy metals, including manganese. Wastewater in the studied section of the river contains no more than 1.5-2 background values of manganese (10 μg/l (Solovov, 1990)).

2.2 Sites And Sampling

The coordinates of the points and their distance from the beginning of the sampling route are given in the table. For convenience, the sampling points are labeled from №1 to № 7. Wastewater is discharged in the river section bounded by sampling points №2 and №3. Thus, of the seven sampling points, two are upstream of such water (points №1 and №2) and the remaining five are downstream, respectively.

Table 1: Sampling points

№ of points	Distance from the start of the route (km)	Terrain coordinates
1	0	48.42837, 135.139195
2	2.2	48.444067, 135.154101
3	4.8	48.451475, 135.176404
4	5.2	48.450723, 135.180436
5	8.6	48.449003, 135.208497
6	18.7	48.476386, 135.308187
7	31.8	48.490070, 135.394574

Samples were collected after the field test of the water quality parameters in 1500 ml plastic bottle and the containers were filled to the total volume and then locked with cap so that no air space can remain inside the bottle. The plastic bottles were first washed thoroughly with soda water, rinsing thoroughly with HNO_3 and distilled water before collecting the sample to make sure that, it is completely free from any undesirable materials according to (APHA, 2017).

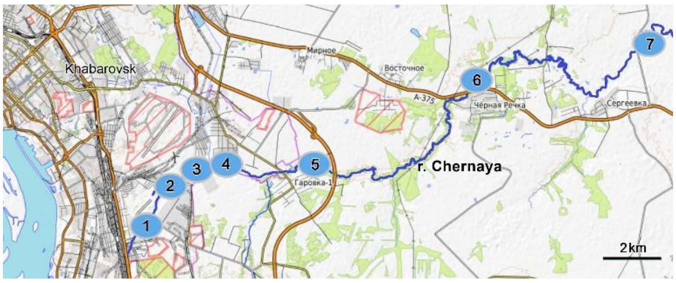


Figure 1: Location map of sampling points from the main channel of the Chernaya River in Khabarovsk and its vicinity

2.3 Hydrochemical And Data Analysis

Manganese concentration was determined using Agilent Technologies atomic absorption spectrometer: SpectrAA 240 FS AA (Crapnell and Banks, 2022).

The chemical composition (NH_3, P_{ind}, O_2) of water was determined in the laboratory of the Collective Use Center of the Institute of Water and Ecologycal Problems of the Far Eastern Branch of the Russian Academy of Sciences using standard methods of hydrochemical analysis (D, 2012). Sampling was carried out in accordance with regulatory and legal documents in the period 2023-2024 (Gost, 2012; Gost, 2022; RD, 1996).

The number of cultured heterotrophic bacteria was determined by the method of limiting dilutions on agarized nutrient medium (fish-peptone agar diluted 10 times). The total dissolved organic matter content, after separation of suspended solids, was estimated by spectrophotometric method at 254 nm (Shimadzu UV-3600) and expressed as spectral absorption coefficient (SAC254, abs. units) (Thomas and Burgess, 2007).

The data were analyzed in STATISTICA 10 complex.

3. Results And Discussion

The results of the monitoring show that the excess of background values of manganese in the studied watercourse is observed throughout the entire observation period, including both spring-summer and fall-winter periods. The increase of manganese content below the main place of wastewater inflow ranged from 3 to 12 times. At the same time, the maximum recorded content was 1198 μg/l. Analysis of the wastewater itself, which induces this release of manganese, showed that it contained no more than 9-17 μg/l manganese.

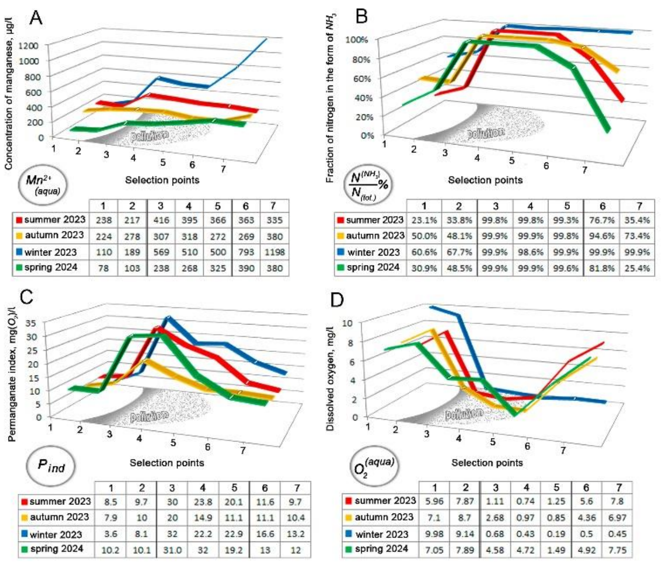
Figure 2A presents data on spatial and temporal dynamics of manganese concentration changes in the waters of the Chernaya River. Figures 2B, 2C, 2D show the hydrochemical parameters that have the greatest influence on manganese migration.

In all presented graphs, we can distinguish 3 plots differing in the dynamics of changes in the registered indicators:

- Site (I), enclosed between points №1 and №2, has a length of 2.2 km. It is characterized by minimal anthropogenic load and relatively stable values of both hydrochemical indicators and manganese content.

- Site (II) is bounded by points №3-№5 and has a length of 3.8 km. It is characterized by the maximum anthropogenic load, since the wastewater inflow is carried out between sampling points №2 and №3. This section shows the greatest change in the values of both manganese content and hydrochemical parameters. Incoming wastewater increases the value of permanganate index in the range from 3 to 11 times, while most of the inorganic nitrogen is present in the ammonium form (more than 90% of N total). Oxidation of incoming organic matter consumes a significant part of dissolved oxygen and its content drops 6-20 times. As a result, this section of the watercourse is characterized by a pronounced reductive hydrochemical environment, which favors the process of manganese reduction and its release from bottom sediments.

- At the last section (III), bounded by points №6-№7, 23.2 km long, stabilization of hydrochemical indicators is noted, their values return to the values typical for the first section. It is important to note that the manganese content in water at this site either does not significantly decrease or continues its growth.



	1	2	3	4	5	6	7
summer 2023	238	217	416	395	366	363	335
autumn 2023	224	278	307	318	272	269	380
winter 2023	110	189	569	510	500	793	1198
spring 2024	78	103	238	268	325	390	380

	1	2	3	4	5	6	7
summer 2023	23.1%	33.8%	99.8%	99.8%	99.3%	76.7%	35.4%
autumn 2023	50.0%	48.1%	99.9%	99.9%	99.8%	94.6%	73.4%
winter 2023	60.6%	67.7%	99.9%	98.6%	99.9%	99.9%	99.9%
spring 2024	30.9%	48.5%	99.9%	99.9%	99.6%	81.8%	25.4%

	1	2	3	4	5	6	7
summer 2023	8.5	9.7	30	23.8	20.1	11.6	9.7
autumn 2023	7.9	10	20	14.9	11.1	11.1	10.4
winter 2023	3.6	8.1	32	22.2	22.9	16.6	13.2
spring 2024	10.2	10.1	31.0	32	19.2	13	12

	1	2	3	4	5	6	7
summer 2023	5.96	7.87	1.11	0.74	1.25	5.6	7.8
autumn 2023	7.1	8.7	2.68	0.97	0.85	4.36	6.97
winter 2023	9.98	9.14	0.68	0.43	0.19	0.5	0.45
spring 2024	7.05	7.89	4.58	4.72	1.49	4.92	7.75

Figure 2: Data of spatial and temporal dynamics of changes in manganese concentration and hydrochemical parameters in the waters of the Chernaya River

Analysis of the data obtained during the winter period shows that their dynamics has a number of principal differences from other observation periods. For example, the dynamics of manganese concentration change shows that at site (I) its content is comparable (or even lower) than at the same site in other observation periods. Further, at site (II) there is an expressive increase in manganese concentration by 2-5 times, and at site (III) there is an even greater increase in its content, which reaches 1198 µg/l. The change of other hydrochemical indicators indicates that a pronounced reductive hydrochemical atmosphere of the aquatic environment is formed in the river section after point №3. Unlike other seasons, the dissolved oxygen content remains very low (0.19-0.68 mg/l) not only directly after the wastewater discharge point, but also downstream. Inorganic nitrogen continues to be in a reduced ammonium form. As a result, such a pronounced reducing environment in the watercourse induces the greatest release of manganese in winter.

Thus, the intensity of manganese release in winter period appears to be the highest. Formation of the maximum registered value occurs due to the increment of manganese content by 1088 µg/l relative to point №1. This increment is associated with the increase of permanganate index values by 28.4 mg O_2/l. It follows that in winter period the increase of permanganate index values for each 10 units additionally releases up to 387 µg Mn.

Analysis of the data obtained in the summer period shows that their greatest dynamics is confined not only to the site of the immediate area of sewage inflow, but also to the last interval of the monitoring area. Discharge of effluents also leads to the formation of a restorative hydrochemical environment. However, in contrast to the winter period, at the last monitoring site (point №7), both the level of water oxygenation (7.80 mg/l) and ammonium nitrogen content (up to 35.4%) return to the initial values (point № 1). Thus, in the summer period the expression of reductive hydrochemical processes is much less. The section of the watercourse where such a reducing environment prevails is much shorter. This leads to the fact that manganese release is not so effective: the maximum recorded concentration of 416 µg/l is 3 times less than the winter one, besides, a weakly expressed trend is formed, leading to its decrease to 335 µg/l. Thus, the release of manganese in the summer period is the least intensive. Formation of the maximum registered value occurs due to the increment of manganese content by 178 µg/l relative to point №1. This increment is associated with the increase of permanganate index values by 21.5 mg O_2/l. It follows that in the summer period the increase of permanganate index values for each 10 units additionally releases up to 83 µg Mn.

Analysis of the data obtained in spring and autumn periods shows that their dynamics is close to the summer period. At the same time, in the spring period the minimum manganese content (78 µg/l) among the registered values is observed. The formation of oxidizing hydrochemical environment is also observed at the last site. In spite of this, in contrast to the summer period of observations, even a weakly expressed trend of manganese content decrease in the spring and fall period is not traced. Thus, a similar release of manganese is observed in the spring and fall periods. Formation of the maximum registered value occurs due to the increment of manganese content by 312 µg/l (spring) and by 187 µg/l (autumn) relative to points №1. This increment is associated with an increase in permanganate index values of 21.8 mg O_2/L (spring) and 12.1 O_2/L (fall). This implies that during the spring and fall periods, an increase in permanganate index values for every 10 units additionally releases 143-155 µg Mn.

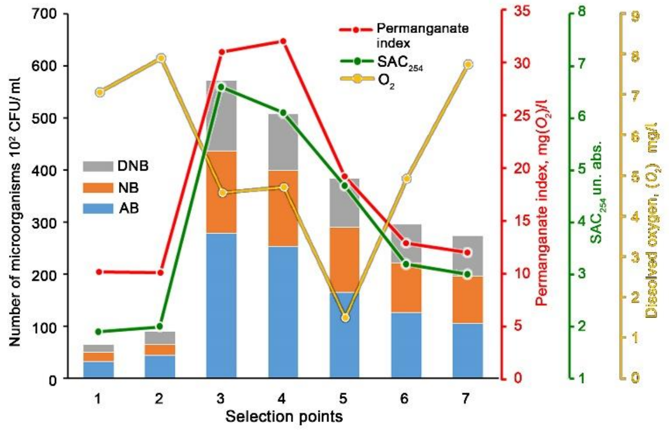


Figure 3: Spatial and temporal dynamics of ammonifying (AB), nitrifying (NB) and denitrifying (DNB) bacterias abundance in spring 2024 and hydrochemical parameters of the samples

For the spring observation period, the microbiome pool of this watercourse was additionally studied (Figure 3). The data of the microbiological study agree well with the previously described results of hydrochemical studies. As expected, the river section downstream of the discharge area shows an expressive increase in microorganisms (MO) abundance. Primarily, the number of MOs increases due to the proportion attributable to primary reducing agents such as ammonifying bacteria (AB). There is then a decline in the number of MOs, first in the AB group and then in other groups. In turn, this follows a decrease in the content of the food resource - organic carbon. The decline in this indicator is observed both from SAC_{254}, units abs. and permanganate index measurements. It is important to note that the processing of primary organic carbon from wastewater by the MOs probably leads to the formation of compounds (products of MO life activity) that contribute to the formation of an even more pronounced reducing environment. This agrees well with the data on the dynamics of oxygen content reduction, when the minimum of its concentration falls on the area following the area of active MO activity. The release of compounds easily involved in reductive processes by microflora may contribute to the increase in the intensity of the release process.

4. CONCLUSIONS

Assessing the toxicological danger of manganese release, we can say that the greatest intoxication for hydrobionts is observed in winter months, when the manganese content reaches more than 1000 µg/L. Manganese release is a consequence of the activation of reductive processes under conditions of pollutant input and simultaneously limited oxygen access due to ice cover. The intensity of manganese release is maximized in this case. On average, in the winter period the increase of permanganate index values for every 10 mg(O_2)/l additionally releases up to 387 µg Mn.

In the absence of ice cover in spring-summer and fall, the intensity of reduction processes decreases. As a result, manganese release occurs with lower intensity. During the summer period, the intensity is minimal, when on average no more than 83 µg Mn is released with an increase in permanganate index for every 10 mg(O_2)/l. During the rest of the warm season (spring and fall), the release of manganese is 143-155 µg for every 10 mg(O_2)/l increase in permanganate index.

It should be noted that the effects of anthropogenic influence on the watercourse are quickly enough leveled in spring, autumn and summer periods. This can be observed at the third section of the sampling route. Characteristic is the decrease of permanganate index value, ammonium nitrogen fraction, as well as total dissolved organic matter content estimated by SAC_{254} value. These data are in good agreement with the dynamics of decrease in the number of ammonifying and nitrifying bacteria.

It is especially important that even after the oxygenation level in the watercourse returns to stable-high values, there is no dynamics of manganese content decrease due to its oxidation to insoluble form MnO_2. This fact is of particular concern, as it demonstrates the possibility of formation of new migration pathways of this element on the territory of this region, initiated by anthropogenic impact.

Acknowledgment

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